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Magnetically controlled electron localization in Eu-rich EuO†

J. Kübler

Abteilung für Physik, Ruhr-Universität Bochum, 463 Bochum, Germany

D. T. Vigren

Institut für Theoretische Physik, Freie Universität Berlin, 1 Berlin 33, Germany

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A calculation of the linear-response properties of an electron gas, coupled by the s-f exchange interaction to a lattice of localized spins is presented. Coupled-charge and spin-density-response functions are determined in the random-phase approximation and used to compute a Mott-type instability of metallic Eu-rich EuO toward formation of magnetic polarons bound to oxygen vacancies. Critical carrier densities are obtained as a function of temperature and phase diagrams of the metallic and insulating states are presented and compared with existing theories and experimental results.

I. INTRODUCTION

An insulator-metal transition (IMT) in Eu-rich EuO has been extensively investigated in recent years.[1-11] The resistivity, found to be nonactivated (metallic) at low temperatures, increases sharply by some orders of magnitude in the vicinity of 50 K. Somewhat above the ferromagnetic Curie temperature of $T_C \simeq 69$ K, the resistivity exhibits a maximum and, depending on sample preparation, shows activated or nonactivated behavior at temperatures much higher than the Curie temperature.[10]

Since the IMT occurs only in Eu-rich EuO, the transition is presumably associated with oxygen vacancies. According to some authors,[1,2] these vacancies act as traps which may or may not bind electrons depending on their position relative to a magnetically shifted conduction-band edge. At the same time it was suggested by von Molnar and Kasuya[8] that the doubly charged positive oxygen vacancies can bind two electrons in a tightly bound nonmagnetic heliumlike singlet in the high-temperature insulating region and in a magnetic triplet below T_C in which the $1s\uparrow$ electron is tightly bound and the $2s\uparrow$ electron merges with the tail of the conduction band edge, giving rise to hopping or even band conduction. In contrast, Torrance *et al.*[9] proposed that the high-temperature state was a "bound magnetic polaron" (BMP); the effect of the deeply trapped electron was ignored and the more loosely bound electron was shown to gain stability in the paramagnetic region by exchange polarizing the Eu^{++} spins within a rather large region about the vacancy.

Other theoretical treatments of the IMT in EuO are those of Nagaev and Grigin[12] and of Leroux-Hugon.[13] Leroux-Hugon, who neglects the effects of double occupancy on vacancy sites, calculates the Mott instability of an exchange-polarized electron gas toward formation of a bound hydrogenic impurity state, using a Landau theory for the local moments and the Thomas-Fermi approximation for the conduction electrons. Nagaev and Grigin compute the linear response functions of an exchange polarized electron gas to an electric perturbation. But, besides neglecting the effects of double occupancy on vacancy sites also, they compute the linear response functions in the spin-wave approximation, which limits their calculation to low temperatures.

In this paper we present a calculation of the electric and magnetic linear-response properties of an electron gas, coupled by the s-f exchange interaction to a lattice of localized spins. Owing to this exchange coupling the electronic charge density, as well as the spin density of the local moments, separately respond to *both* electric and magnetic field perturbations. This is an exact property within linear response and can be formulated as

$$\begin{aligned} \delta\langle\rho\rangle &= L_{11}\varphi + L_{12}h\,,\\ \delta\langle S^z\rangle &= L_{21}\varphi + L_{22}h\,, \end{aligned} \tag{1}$$

where $\delta\langle\rho\rangle$ and $\delta\langle S^z\rangle$ are the charge and spin densities, respectively, induced by the electric and magnetic perturbations, φ and h, respectively. In Sec. II the response functions L_{11}, L_{12}, L_{21}, and L_{22} are calculated using the Green's-function equation of motion method and decoupling in the random-phase approximation[14] (RPA). In Sec. III we use these response functions to calculate the Mott instability of a conduction electron toward formation of a bound magnetic polaron in the field of a positive oxygen vacancy and interacting via s-f exchange with the localized magnetic moments of the Eu^{++} ions. Since the vacancy is doubly charged, we assume a second electron to be tightly bound to it. Although this second electron does not take part in the conduction process, it interacts with the localized magnetic moments of the nearest Eu^{++} neighbors and gives rise to a perturbing nonuniform molecular field that sensitively influences the Mott localization of the conduction electrons, even well above T_C. Our approach is similar to those of Nagaev and Grigin,[12] Leroux-Hugon,[13] and to the model of Torrance *et al.*[9] However, we believe, in contrast to these authors, that the effects of the tightly bound electron cannot be ignored. Further, our RPA treatment avoids some approximations and limitations and exhibits the effects that we believe are responsible for the IMT. A discussion of the results is given in Sec. IV.

II. LINEAR-RESPONSE THEORY

We consider a system of localized magnetic moments arranged on a crystal lattice and interacting via the Heisenberg Hamiltonian

$$H_f = \frac{1}{2} \sum_{n,m} J_{nm} \vec{S}_n \cdot \vec{S}_m + g\mu_B h_0 \sum_n S_n^z , \qquad (2)$$

where $\vec{S}_n$ is the spin moment on the nth site and J_{nm} is the exchange coupling constant between the sites n and m. The quantity g is the gyromagnetic ratio and μ_B is the Bohr magneton. The applied field $\vec{h}_0$ serves to define the direction $\vec{z}$ of spontaneous magnetization. Equation (2) describes the magnetic subsystem, which we take to be immersed in an interacting sea of conduction electrons described by the Hamiltonian

$$H_s = \sum_{\vec{p},\sigma} E_{\vec{p}}\, a^\dagger_{\vec{p}\sigma} a_{\vec{p}\sigma} + \frac{1}{2} \sum_{\substack{\vec{p},\vec{q},\vec{K}\\ \sigma,\sigma'}} V_K\, a^\dagger_{\vec{q}-\vec{K}\sigma} a^\dagger_{\vec{p}+\vec{K}\sigma'} a_{\vec{p}\sigma'} a_{\vec{q}\sigma} , \qquad (3)$$

where $E_{\vec{p}}$ is the conduction-band energy as calculated in the paramagnetic state and is assumed to be a simple s band. The quantity

$$V_K = 4\pi e^2/\kappa_0 K^2$$

is the Fourier transform of the Coulomb interaction between the conduction electrons in a medium of unit volume, described by the background dielectric constant κ_0. The quantities $a^\dagger_{\vec{p}\sigma}$ and $a_{\vec{p}\sigma}$ are Bloch creation and annihilation operators. Equation (3) describes the electronic subsystem. The magnetic and electronic subsystems are coupled by the s-f Hamiltonian[15]

$$H_{sf} = -I \sum_{n,\sigma,\sigma'} c^\dagger_{n\sigma} \vec{\sigma}_{\sigma\sigma'} \cdot \vec{S}_n c_{n\sigma'} , \qquad (4)$$

where $c^\dagger_{n\sigma}$ and $c_{n\sigma}$ are Wannier creation and annihilation operators related to the corresponding Bloch operators by means of

$$a^\dagger_{\vec{p}\sigma} = \frac{1}{\sqrt{N}} \sum_n e^{i\vec{p}\cdot\vec{R}_n} c^\dagger_{n\sigma} \qquad (5)$$

and its Hermitian conjugate. The quantity $\vec{\sigma}$ is the Pauli spin operator, I is the contact intra-atomic s-f exchange parameter, and N is the number of atomic sites.

The total system under consideration is then described by

$$H_0 = H_f + H_s + H_{sf} . \qquad (6)$$

We now apply, adiabatically, small electric and magnetic fields whose time and spatial variations are characterized by a frequency ω and a wave vector $\vec{k}$. The corresponding perturbation is denoted by H_1 and

$$H_1 = H_m + H_e , \qquad (7)$$

where

$$H_m = g\mu_B N h(\vec{k},\omega) S^z_{\vec{k}} e^{-i\omega t + \eta t} + \text{H.c.} \qquad (8)$$

and

$$H_e = -e\rho^\dagger_{\vec{k}} \varphi(\vec{k},\omega) e^{-i\omega t + \eta t} + \text{H.c.} \qquad (9)$$

Here $h(\vec{k},\omega)$ and $\varphi(\vec{k},\omega)$ are the Fourier components of the perturbing magnetic field and electric potential. The spatial distributions of spin and electron densities are determined by $S^z_{\vec{k}}$ and $\rho^\dagger_{\vec{k}}$ which may be written

$$S^z_{\vec{k}} = \frac{1}{N} \sum_m e^{i\vec{k}\cdot\vec{R}_m} S^z_m \qquad (10)$$

and

$$\rho^\dagger_{\vec{k}} = \sum_{\vec{p},\sigma} a^\dagger_{\vec{p}\sigma} a_{\vec{p}-\vec{k}\sigma} = \rho_{-\vec{k}} . \qquad (11)$$

To formulate the response properties of the system defined by (6), we use the linear response formula[16]

$$\delta A = \int_{-\infty}^{\infty} \langle\langle A(t-t'); H_1(t') \rangle\rangle \, dt' . \qquad (12)$$

Here δA represents the response of the quantity A to a time-dependent perturbation $H_1(t)$. It is just the difference in the thermal average of A computed with and without $H_1(t)$ applied. The retarded double-time Green's function, whose propagation is determined by the unperturbed Hamiltonian H_0, appears under the time integral and the quantity $A(t)$ is the operator A expressed in the Heisenberg representation. Specifying this operator to be either $\rho_{\vec{k}}$ defined by (11) or $S^z_{\vec{k}}$ defined by (10) and substituting for H_1 [Eq. (7)], we obtain for the time-Fourier transforms of the longitudinal charge- and spin-density response

$$\delta\rho_{\vec{k}}(\omega) = L_{11}(\vec{k},\omega)\varphi(\vec{k},\omega) + L_{12}(\vec{k},\omega)h(\vec{k},\omega), \tag{13}$$

$$\delta S^z_{\vec{k}}(\omega) = L_{21}(\vec{k},\omega)\varphi(\vec{k},\omega) + L_{22}(\vec{k},\omega)h(\vec{k},\omega)$$

and find the response coefficients to be given by

$$L_{11}(\vec{k},\omega) = -2\pi e\langle\langle \rho_{\vec{k}};\rho_{-\vec{k}}\rangle\rangle_\omega, \tag{14}$$

$$L_{12}(\vec{k},\omega) = 2\pi N g\mu_B\langle\langle \rho_{\vec{k}};S^z_{\vec{k}}\rangle\rangle_\omega, \tag{15}$$

$$L_{21}(\vec{k},\omega) = -2\pi e\langle\langle S^z_{-\vec{k}};\rho_{-\vec{k}}\rangle\rangle_\omega, \tag{16}$$

$$L_{22}(\vec{k},\omega) = 2\pi N g\mu_B\langle\langle S^z_{-\vec{k}};S^z_{\vec{k}}\rangle\rangle_\omega\ . \tag{17}$$

Here the ω subscript of the Green's functions is used to denote the time-Fourier transforms and is usually suppressed but implied.

To actually calculate the response coefficients, we must resort to approximations: The Green's functions are calculated by the Zubarev equations of motion method[16] and are decoupled in the RPA.[17] Furthermore, in the calculation we use only the Zener part[15] of Eq. (4) since spin-flip terms in H_{sf} give rise to mathematical difficulties. If, however, one attempts to determine the spin-flip Green's functions, one finds that some of them renormalize the Heisenberg coupling constants J_{nm} producing indirect-exchange contributions. We therefore reinterpret the coupling constants J_{nm} in the final answers to be renormalized, although this does not account for all the neglected terms. We shall find another coupling constant later on, j defined by Eq. (55), which we also find necessary to renormalize, presumably for the same reasons.

For the detailed calculation it is convenient to define up- or down-spin charge density operators $\rho_{\vec{k}\sigma}$ by

$$\rho_{\vec{k}\sigma} = \sum_{\vec{p}} a^\dagger_{\vec{p}\sigma} a_{\vec{p}+\vec{k}\sigma}\ . \tag{18}$$

It is then easily seen that for the determination of Eqs. (14)–(17) three Green's functions are needed. Setting up their equations of motion and decoupling, one obtains the set of equations

$$\langle\langle \rho_{\vec{k}\sigma};\rho_{-\vec{k}}\rangle\rangle = -\chi_{0\sigma}(\vec{k},\omega)[1/2\pi + V_k\langle\langle \rho_{\vec{k}};\rho_{-\vec{k}}\rangle\rangle + I(\delta_{\sigma,\downarrow}-\delta_{\sigma,\uparrow})\langle\langle \rho_{\vec{k}};S^z_{\vec{k}}\rangle\rangle], \tag{19}$$

$$\langle\langle \rho_{\vec{k}\sigma};S^z_{\vec{k}}\rangle\rangle = -\chi_{0\sigma}(\vec{k},\omega)[V_k\langle\langle \rho_{\vec{k}};S^z_{\vec{k}}\rangle\rangle + I(\delta_{\sigma,\downarrow}-\delta_{\sigma,\uparrow})\langle\langle S^z_{-\vec{k}};S^z_{\vec{k}}\rangle\rangle], \tag{20}$$

$$\langle\langle S^z_{-\vec{k}};S^z_{\vec{k}}\rangle\rangle = \frac{-\chi^H_{zz}(\vec{k},\omega)}{2\pi N(g\mu_B)^2} \times[1-2\pi I(\langle\langle \rho_{\vec{k}\uparrow};S^z_{\vec{k}}\rangle\rangle - \langle\langle \rho_{\vec{k}\downarrow};S^z_{\vec{k}}\rangle\rangle)]\ . \tag{21}$$

Here $\chi_{0\sigma}(\vec{k},\omega)$ is the Lindhard susceptibility function[17]

$$\chi_{0\sigma}(\vec{k},\omega) = \sum_{\vec{p}} \frac{f_{\vec{p}+\vec{k}\sigma} - f_{\vec{p}\sigma}}{\omega - E_{\vec{p}+\vec{k}} + E_{\vec{p}}}\ ; \tag{22}$$

$f_{\vec{p}\sigma}$ is the Fermi-Dirac distribution

$$f_{\vec{p}\sigma} = \{\exp[\beta(E_{\vec{p}} \pm SI\sigma_0 - \mu)]+1\}^{-1}\ , \tag{23}$$

where μ is the chemical potential $\beta = 1/(k_BT)$; and σ_0 is the reduced magnetization due to the lattice of localized moments with spin S. The quantity $\chi^H_{zz}(\vec{k},\omega)$ is given by

$$\chi^H_{zz}(\vec{k},\omega) = F_1(\vec{k},\omega)/[\omega - F_2(\vec{k},\omega)]\ , \tag{24}$$

where

$$F_1(\vec{k},\omega) = -\frac{(g\mu_B)^2}{2}\sum_{\vec{p}}\frac{(\psi_{\vec{p}}-\psi_{\vec{p}+\vec{k}})(J_{\vec{p}}-J_{\vec{p}+\vec{k}})}{\omega - S\sigma_0(J_{\vec{p}}-J_{\vec{p}+\vec{k}})} \tag{25}$$

and

$$F_2(\vec{k},\omega) = \frac{1}{2}\sum_{\vec{p}}\frac{(J_{\vec{p}}-J_{\vec{p}+\vec{k}})[(J_{\vec{p}}-J_{\vec{k}})\psi_{\vec{p}} + (J_{\vec{k}}-J_{\vec{p}+\vec{k}})\psi_{\vec{p}+\vec{k}}]}{\omega - S\sigma_0(J_{\vec{p}}-J_{\vec{p}+\vec{k}})}\ . \tag{26}$$

Here $J_{\vec{p}}$ is the Fourier transform of J_{nm} and $\psi_{\vec{p}}$ is defined through

$$\langle S^+_{\vec{p}} S^-_{\vec{q}}\rangle = \delta_{\vec{p},\vec{q}}\psi_{\vec{p}}\ .$$

Equations (19)–(21) may be easily solved for the three types of Green's functions and the response coefficients may then be written explicitly as

$$L_{11}(\vec{k},\omega) = e\left(\chi^+_0(\vec{k},\omega) - \frac{4I^2\chi^H_{zz}(\vec{k},\omega)}{N(g\mu_B)^2} \times\chi_{0\uparrow}(\vec{k},\omega)\chi_{0\downarrow}(\vec{k},\omega)\right)\Big/\ \xi(\vec{k},\omega), \tag{27}$$

$$L_{12}(\vec{k},\omega) = \frac{I\chi^H_{zz}(\vec{k},\omega)\chi^-_0(\vec{k},\omega)}{g\mu_B\xi(\vec{k},\omega)}\ , \tag{28}$$

$$L_{21}(\vec{k},\omega) = \frac{-eI\chi^H_{zz}(\vec{k},\omega)\chi^-_0(\vec{k},\omega)}{(g\mu_B)^2N\xi(\vec{k},\omega)}\ , \tag{29}$$

$$L_{22}(\vec{k},\omega) = \frac{-\chi^H_{zz}(\vec{k},\omega)[1+V_k\chi^+_0(\vec{k},\omega)]}{g\mu_B\xi(\vec{k},\omega)}\ , \tag{30}$$

where

$$\chi^\pm_0(\vec{k},\omega) = \chi_{0\uparrow}(\vec{k},\omega) \pm \chi_{0\downarrow}(\vec{k},\omega) \tag{31}$$

and

$$\xi(\vec{k},\omega)=1+V_k\chi_0^+(\vec{k},\omega)-\frac{I^2\chi^H_{zz}(\vec{k},\omega)}{N(g\mu_B)^2}\times[\chi_0^+(\vec{k},\omega)+4V_k\chi_{0\uparrow}(\vec{k},\omega)\chi_{0\downarrow}(\vec{k},\omega)] . \tag{32}$$

Using Eq. (13) one can express the longitudinal dielectric function[17] $\epsilon(\vec{k},\omega)$ and the longitudinal magnetic-susceptibility function[18] $\chi_{zz}(\vec{k},\omega)$, in terms of L_{11} and L_{22}:

$$\epsilon(\vec{k},\omega)=[1-(V_k/e)L_{11}(\vec{k},\omega)]^{-1} \tag{33}$$

and

$$\chi_{zz}(\vec{k},\omega)=-g\mu_B L_{22}(\vec{k},\omega) . \tag{34}$$

Substituting Eqs. (27) and (30), we obtain

$$\epsilon(\vec{k},\omega)=1+V_k\left(\chi_0^+(\vec{k},\omega)-\frac{4I^2\chi^H_{zz}(\vec{k},\omega)}{N(g\mu_B)^2}\chi_{0\uparrow}(\vec{k},\omega)\chi_{0\downarrow}(\vec{k},\omega)\right)\times\left(1-\frac{I^2\chi^H_{zz}(\vec{k},\omega)}{N(g\mu_B)^2}\chi_0^+(\vec{k},\omega)\right)^{-1} \tag{35}$$

and

$$\chi_{zz}(\vec{k},\omega)=\frac{\chi^H_{zz}(\vec{k},\omega)[1+V_k\chi_0^+(\vec{k},\omega)]}{\xi(\vec{k},\omega)} . \tag{36}$$

The basic equations which yield these results are Eqs. (19)–(21). These were obtained by dropping spin-flip Green's functions of the type

$$\langle\langle S^-_{\vec{k}}a^\dagger_{\vec{k}\uparrow}a_{\vec{k}\downarrow};S^z_{\vec{k}}\rangle\rangle$$

and

$$\langle\langle S^-_{\vec{p}-\vec{q}}a^\dagger_{\vec{p}\uparrow}a_{\vec{q}+\vec{k}\downarrow};\rho_{\vec{k}}\rangle\rangle$$

and decoupling the hierarchy of Green's functions as follows[19]:

$$\langle\langle a^\dagger_{\vec{p}s}\rho_{\vec{k}}a_{\vec{q}s'};\rho_{-\vec{k}}\rangle\rangle\simeq\langle a^\dagger_{\vec{p}s}a_{\vec{q}s'}\rangle\langle\langle\rho_{\vec{k}};\rho_{-\vec{k}}\rangle\rangle+\langle\rho_{\vec{k}}\rangle\langle\langle a^\dagger_{\vec{p}s}a_{\vec{q}s'};\rho_{-\vec{k}}\rangle\rangle ,$$

$$\langle\langle S^+_{\vec{p}}S^z_{\vec{q}}S^-_{\vec{r}};S^z_{\vec{k}}\rangle\rangle\simeq\langle S^+_{\vec{p}}S^-_{\vec{r}}\rangle\langle\langle S^z_{\vec{q}};S^z_{\vec{k}}\rangle\rangle+\langle S^z_{\vec{q}}\rangle\langle\langle S^+_{\vec{p}}S^-_{\vec{r}};S^z_{\vec{k}}\rangle\rangle .$$

To show that this decoupling scheme corresponds to the RPA, we set $I=0$ in Eqs. (35) and (36) and obtain

$$\epsilon(\vec{k},\omega)=1+V_k\chi_0^+(\vec{k},\omega) , \tag{37}$$

$$\chi_{zz}(\vec{k},\omega)=\chi^H_{zz}(\vec{k},\omega) . \tag{38}$$

Equation (37) is the standard RPA result[17] for the longitudinal dielectric function of the electron gas. The quantity $\chi^H_{zz}(\vec{k},\omega)$ is seen to be the longitudinal magnetic susceptibility of the Heisenberg-spin subsystem. If we take the $\omega\to 0$ limit of Eq. (24), and assume $S=\frac{1}{2}$ so that in the Tyablikov theory we may write[20]

$$\psi_{\vec{p}}=\frac{\sigma_0}{N}\{\exp(\beta[g\mu_B h_0+\tfrac{1}{2}\sigma_0(J_0-J_{\vec{p}})])-1\}^{-1} , \tag{39}$$

we obtain $\chi^H_{zz}(\vec{k},0)$ which agrees with the results of Kawasaki and Mori[21] and of Liu.[22] These authors use essentially the Tyablikov approximation which is the RPA for spin systems.

For use in Sec. III one may determine the integrals occuring in Eqs. (25) and (26) in the limit $\omega\to 0$ and for long wavelengths, where

$$J_0-J_{\vec{p}}=c_J p^2 .$$

For $T<T_C$ one then obtains[21]

$$\chi^H_{zz}(\vec{k},0)=\frac{(g\mu_B)^2}{c_J}\left[k^2+k2c_J N/k_B T\left(1-\frac{d\ln\sigma_0}{d\ln T}\right)\right]^{-1} , \tag{40}$$

where N is here the number of spins per unit volume. For $T>T_C$ the Ornstein-Zernike result is obtained.[23]

From Eqs. (28) and (29) one sees that the off-diagonal response functions, L_{12} and L_{21}, vanish as expected for $I=0$. They also vanish when the exchange splitting of the conduction band goes to zero (i.e., for $T>T_C$). In this case $\chi_{0\uparrow}=\chi_{0\downarrow}=\frac{1}{2}\chi_0^+$ and $\chi_0^-=0$. Also for $T>T_C$ the dielectric function reduces to the RPA result, Eq. (37), and the longitudinal susceptibility reduces to

$$\chi_{zz}(\vec{k},\omega)=\chi^H_{zz}(\vec{k},\omega)\left(1-\frac{I^2\chi^H_{zz}(\vec{k},\omega)}{N(g\mu_B)^2}\chi_0^+(\vec{k},\omega)\right)^{-1} . \tag{41}$$

In the static limit, $\omega=0$, and for small k, this can easily be reduced to the Ornstein-Zernike susceptibility,[23] except for a renormalization of T_C due to the s-f exchange coupling.

Finally, it should be noted that for $T<T_C$ in the case that one subband is populated (i.e., $\chi_{0\downarrow}=0$), if we replace $\chi^H_{zz}(\vec{k},0)$ by the Ornstein-Zernike susceptibility, our response functions ϵ and χ_{zz} reduce to the results of Leroux-Hugon[13] in the long-wavelength static limit.

III. INSULATOR-METAL TRANSITION IN EuO

We want to apply our results to the IMT in Eu-rich ferromagnetic EuO. In view of modern theories concerning localization in disordered systems,[24] one ought to study Anderson localization of electrons in the random field of oxygen vacancies which according to all experimental indications are responsible for the IMT in EuO. Such a theory, being complicated in nonmagnetic systems, is expected to be even more difficult here because of the magnetic properties of EuO that manifestly influence the IMT.

Therefore, we use the older idea of Mott[25] to calculate the critical carrier density n_c, that separates activated (semiconducting) from non-activated (metallic) behavior in systems with charged impurities. This theory is not only easily applied, and modified for the magnetic properties of the system, but estimates of n_c for nonmagnetic-doped semiconductors have ordinarily been found to be surprisingly accurate.[26]

The idea of Mott localization about charged impurities can be summarized as follows: In the metallic phase, charged impurities are screened and can or cannot bind an electron depending on the carrier density. If binding is possible, the assumed metallic state is unstable and the system is semiconducting provided the number of impurities is large enough. Within a simple Thomas-Fermi theory one finds the condition[27]

$$a_0^2 k_s^2 = 1 \tag{42}$$

determines the critical density. Here a_0 is the crystal Bohr radius and k_s is the Thomas-Fermi inverse screening length,

$$k_s^2 = 6\pi e^2 n / (\kappa_0 E_F),$$

where κ_0 is the background dielectric constant and E_F is the Fermi energy

$$E_F = (\hbar^2/2m^*)(3\pi^2 n)^{2/3}\ , \tag{43}$$

where n is the carrier density and m^* is the effective mass. Equation (42) then gives for the critical carrier density

$$a_0 n_c{}^{1/3} \simeq 0.25\ . \tag{44}$$

For $n < n_c$ the system can be semiconducting. In ferromagnetic EuO, however the conduction band is known to be split for $T < T_C$ due to the long-range magnetic order and the s-f exchange interaction. The splitting is so large that for carrier densities of the order of 10^{19} cm^{-3} only the spin-up band is populated at low temperatures. Consequently, Eq. (43) becomes for $T \ll T_C$

$$E_F = (\hbar^2/2m^*)(6\pi^2 n)^{2/3}\ . \tag{45}$$

This leads to the relation

$$n_c^{\mathrm{EuO}} = 4 n_c^{\mathrm{nonmagn}} \tag{46}$$

where n_c^{nonmagn} is calculated with Eq. (44). Hence, if the carrier density is larger than n_c^{nonmagn} but smaller than $4n_c^{\mathrm{nonmagn}}$, the system can be nonmetallic at low temperatures and metallic at high temperatures, the transition occuring where the long-range magnetic order disappears, i.e., at or close to T_C. A more accurate calculation, in particular, a better theory for screening, will change the numerical estimates somewhat, but will not change the general situation, which is shown as curve A in Fig. 1.

Therefore, according to this consideration, which takes into account the magnetic order only through the exchange splitting of the conduction band, if a phase change occurs at all, then the high-temperature phase should be metallic and the low-temperature phase should be insulating, contrary to the experimental observation on Eu-rich EuO. If, however, the dielectric screening is computed with $\epsilon(\vec{k}, 0)$ given by Eq. (35), and if the magnetic interaction of charge carriers with the local moments is taken explicitly into account, an IMT with a low-temperature metallic phase may be obtained over a definite range of concentrations.

To show this we proceed by assuming a metallic system with n charge carriers per cm^3. An equal amount of doubly charged positive oxygen vacancies is assumed to have trapped one electron each in a tightly bound state. We compute whether such an impurity complex, composed of vacancy and tightly bound electron, is capable of binding a second electron in the form of a bound-magnetic polaron[9,13] (BMP). If the result is affirmative, then the metallic state is unstable toward formation of a BMP. We should emphasize that no attempt is made to calculate or describe the system after localization occurs. Rather our purpose is to determine $n_c(T)$, which gives the phase boundary of the metallic state.

The trial wave function of the " second" electron

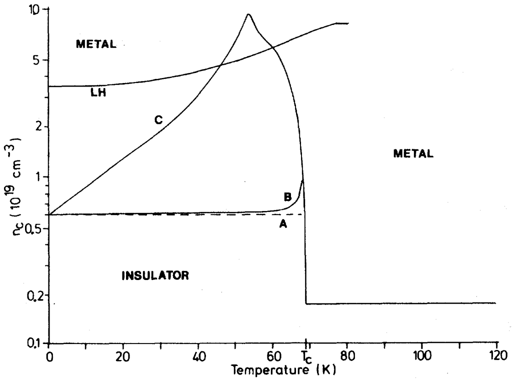

FIG. 1. Critical carrier density as a function of temperature. Curve A: normal Mott transition with exchange split bands below $T_C = 69$ K. Curve B: Mott instability toward formation of a BMP using the Ornstein-Zernike susceptibility in the screening function and $\eta = 0$ in Eq. (52). Curve C: Same as B, but using the Kawasaki-Mori instead of the Ornstein-Zernike susceptibility. Curve LH: reproduced here for comparison from Ref. 13.

is written

$$\Psi=(\pi a^3)^{-1/2}e^{-r/a}\ . \tag{47}$$

This electron interacts with the doubly charged screened vacancy, with the Eu^{++} spins, and with the tightly bound electron. The latter interaction gives rise to Coulomb and exchange terms, as in the helium problem. If Ψ is weakly bound, the Coulomb term just cancels one charge of the vacancy and the exchange term proves to be negligible. Neglecting the interaction with the other conduction electrons, we obtain for those contributions to the energy that depend on the variational parameter a

$$E=\frac{\hbar^2}{2m^*a^2}-\frac{32e^2}{\pi a^4\kappa_0}\int_0^\infty\frac{dk}{\epsilon(k,0)(k^2+4/a^2)^2}+\frac{I}{(2\pi)^3}\int Q_{\vec{k}}\,\delta S^z_{\vec{k}}\,d^3k, \tag{48}$$

where Q_k is the Fourier transform of $|\Psi|^2$,

$$Q_{\vec{k}}=(16/a^4)(k^2+4/a^2)^{-2}\ . \tag{49}$$

The first term in Eq. (48) is the kinetic energy, the second is the potential energy in the field of the screened vacancy where one charge has been canceled by the tightly bound electron. It is the third term that lead us to call the electron described by Ψ a "magnetic polaron" since it arises from the exchange binding due to the local spin polarization $\delta S^z_{\vec{k}}$. There are two effects that give rise to the local spin polarization. One is the electric potential of the vacancy which induces a spin polarization $\delta S^z_{\vec{k}}$, that is to be obtained from the second of the linear response formulas given by Eq. (13), with

$$\varphi(\vec{k},0)=4\pi e/\kappa_0 k^2\ . \tag{50}$$

The other effect that gives rise to local spin polarization is the localized charge and spin density due to the electron Ψ itself. According to Yanase and co-workers[28] it can be described by an effective molecular field which must be determined self-consistently. Under certain circumstances this field can cause a magnetic polaron to form near the Curie temperature and will lead to self-trapping of charge carriers. Its contribution to $\delta S^z_{\vec{k}}$ could be obtained from the second of the linear response formulas given by Eq. (13) by an appropriate choice of $h(\vec{k},0)$.

However, the vacancy gives rise to another nonuniform magnetic field of exchange origin. This occurs because the tightly bound electron interacts with the Eu^{++} ions near the vacancy causing a Eu^{++} spin cluster to form. The interaction energy is of the form

$$\sum_n \tilde{J}_{0n}\vec{S}_n\cdot\vec{s}_0\ ,$$

where $\vec{S}_n$ are the localized Eu^{++} spins, $\vec{s}_0$ is the spin of the tightly bound electron in the vacancy, the sum extends over the spin cluster, and $\tilde{J}_{0n}$ is an exchange coupling constant which is of the order of the s-f interaction I. Since $IS\simeq 0.1$ eV, the spin cluster may persist far above T_C in a nearly saturated configuration. From a mean-field viewpoint, the spin cluster breaks the translational symmetry of the spin system, giving rise to a nonuniform perturbation of the Weiss molecular field in the neighborhood of the vacancy. At low temperatures, when the total spin system approaches saturation, the spatial uniformity of the effective field must be restored. We represent these effects by writing

$$h(\vec{r})=\tilde{J}e^{-\gamma r}(1-\sigma_0)/g\mu_B\ , \tag{51}$$

where $h(\vec{r})$ represents the spatial deviation from a uniform field. The exponential factor reflects that nonuniformity exists only near the vacancy and the factor $(1-\sigma_0)$ accounts for the restoration of spatial uniformity when the Eu^{++} spins approach saturation $(\sigma_0\rightarrow 1)$. Above T_C, $h(\vec{r})$ will fluctuate with a time characteristic of local spin relaxation. But this time is expected to be much larger than that characterizing the electronic relaxation so that electrons see an essentially constant perturbation near the vacancy. At very high temperatures such fluctuations will become important, however, and $h(\vec{r})$ will become a rapidly decreasing function of temperature. In our treatment we neglect this effect and treat $h(\vec{r})$ as a function independent of temperature.

We believe the molecular field due to the tightly bound electron, Eq. (51), to dominate the Yanase molecular field, which has been shown to give rise to self-trapping only in a few rare cases that are not likely to be realizable in the Eu chalcogenides.[28] We therefore neglect its effect.

It is now easy to substitute the second of Eqs. (13) into (48), using the Fourier transform of (51) for $h(\vec{k})$, $\varphi(\vec{k})$ as given by (50), and the response coefficients L_{21} and L_{22} as given by (29) and (30). The result can be expressed in dimensionless form by dividing with the crystal Rydberg

$$E_0=\hbar^2/(2m^*a_0^2),$$

where a_0 is the crystal Bohr radius. The quantity to be minimized can then be written

$$\bar{E} \equiv \frac{E}{E_0} = \alpha^2 - 64 a_0 \alpha^4 \pi^{-1}$$
$$\times \left(\int_0^\infty \frac{[1 - 2I^2 \chi_{zz}^H(k,0) \chi_{0\downarrow}(k,0)/(Ng^2\mu_B^2)]\, dk}{\xi(k,0)(a_0^2k^2 + 4\alpha^2)^2} + \eta(1-\sigma_0) \int_0^\infty \frac{k^2 \chi_{zz}(k,0)}{(a_0^2k^2 + a_0^2\gamma^2)^2 (a_0^2k^2 + 4\alpha^2)^2} dk \right) . \tag{52}$$

The functions $\xi(k,0)$, $\chi_{zz}^H(k,0)$, $\chi_{0\sigma}(k,0)$, and $\chi_{zz}(k,0)$ are defined in Sec. II by the Eqs. (32), (40), (22), and (36), respectively, and α is the variational parameter $\alpha = a_0/a$. The quantity η is given by

$$\eta = I \tilde{J} a_0^3 \gamma / 2E_0 g^2 \mu_B^2 . \tag{53}$$

The variational $\bar{E}$ is numerically minimized; if $\bar{E} \geq 0$ the metallic phase is stable; if $\bar{E} < 0$ the conduction electrons become localized in helium-like impurity states, whose actual ground state, however, we cannot infer from our calculation.

To establish contact with Leroux-Hugon's calculations,[13] we at first ignore the magnetic field perturbation and set $\eta = 0$ in Eq. (52). Further, we replace $\chi_{zz}^H(k,0)$ by the Ornstein-Zernike susceptibility, $\chi^{OZ}(k)$ which for $T < T_C$ can be written[23]

$$\chi^{OZ}(k) = \frac{(g\mu_B)^2}{c_J} \left(k^2 + \frac{2a'}{c_J}(T_C - T) \right)^{-1} , \tag{54}$$

where $a' = 3k_B/S(S+1)$, k_B being Boltzmann's constant. All terms in Eq. (52) involving I^2 can be combined to give a factor

$$j = (IS)^2 \kappa_0 / 4\pi N e^2 c_J , \tag{55}$$

where $c_J = 2Jl^2$ for a fcc ferromagnet of lattice constant l and nearest-neighbor exchange constant J. In the notation of Ref. 13, $j = \beta E_i^2$. Using, for κ_0, the static dielectric constant of EuO, $\kappa_0 = 23.9$,[29] $IS = 0.26$ eV,[30,31] $l = 5.15$ Å,[32] and $J = 0.58 \times 10^{-4}$ eV,[32] we obtain $j \simeq 90$. This value is much too large, implying, in contrast to the conclusions of Ref. 13, that divergences in the response functions are expected below T_C which indicate instabilities in the translationally invariant electronic and magnetic systems. We therefore reinterpret j to be an *effective* coupling constant and carry out the calculation with $j = 0.9$. We return to this point in Sec. IV. The result of this calculation is shown as curve B in Fig. 1 which gives the critical carrier density as a function of temperature. An effective mass $m^* = 0.55 m_0$ was assumed and, for simplicity, a simple molecular field magnetization was used for σ_0. This curve should be compared with the curve labeled *LH*, which is taken from Ref. 13. Owing to Leroux-Hugon's choice of $m^*/m_0 = 1$, his critical-carrier concentration is higher at $T = 0$ than ours. The different shape is due to his different estimate of the other numerical constants. Furthermore, in Ref. 13 it was assumed that only the spin-up band was populated, so that n_c shows no tendency near T_C to decrease to the paramagnetic value (which should be $\frac{1}{4}$ times the $T = 0$ value as discussed above.).

In curve C of Fig. 1, $\chi_{zz}^H(k,0)$ given by Eq. (40) is used, again assuming a simple molecular-field magnetization for σ_0. The same numerical constants were used as in curve B and magnetic effects of the impurity as well as self-trapping effects were ignored (i.e., $\eta = 0$). At $T = 0$, for a carrier density of $n = 3 \times 10^{19}$ cm^{-3}, our calculation predicts metallic behavior for temperatures up to 40 K; between 40 and 66 K, the system is found to be insulating; above 66 K the metallic phase is again stable. Systems with a carrier density in excess of $n \simeq 0.17 \times 10^{19}$ cm^{-3} should be metallic in the paramagnetic state. No phase change is expected for carrier densities in excess of $n \simeq 10^{20}$ cm^{-3}.

The curves of Fig. 2 show the effect of the magnetic perturbation, given by Eq. (51). Using the same numerical constants as in Fig. 1 with $\tilde{J} = 0.027 I$ and $a_0\gamma = 4$, we obtain curve A and with $\tilde{J} = 0.12 I$ and $a_0\gamma = 3.15$, we obtain curve B. At low temperatures the situation is qualitatively similar to the case $\eta = 0$. Above T_C, however, the insulating phase may persist far into the paramagnetic region. In the case of curve B, a system with a carrier density of $n = 3 \times 10^{19}$ cm^{-3}, is metallic up to about 30 K, but then insulating up to ~100 K.

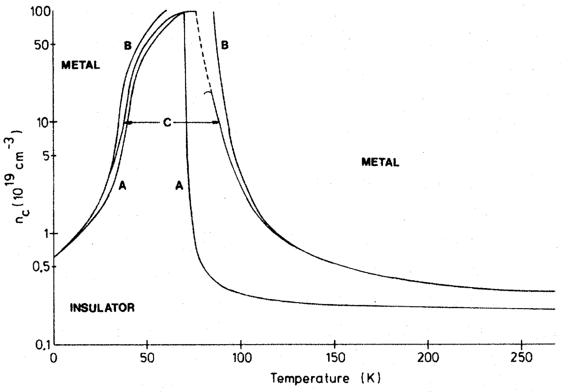

FIG. 2. Critical carrier density as a function of temperature. Mott instability toward formation of a BMP using the Kawasaki-Mori susceptibility in the screening function and including effects of the tightly bound electron. Curve A: $\tilde{J} = 0.027\, I$ and $a_0\gamma = 4$ in Eq. (52). Curve B: $\tilde{J} = 0.12\, I$ and $a_0\gamma = 3.15$. Curve C: same as B but now an external magnetic field of 10 kOe is applied. (Dashed portion: interpolation near T_C to indicate expected behavior.)

Curve C in Fig. 2 shows the effect of an externally applied magnetic field; it was computed with the parameters of curve B and an external field of $H_{\text{ext}} = 10$ kOe. The IMT at low temperatures is seen to be shifted to a higher temperature, the shift being larger for larger carrier densities. (The dashed portion was drawn near T_C, where numerical results became unreliable, to indicate the expected behavior.)

IV. DISCUSSION

We have calculated the longitudinal charge- and spin-density response of a system of conduction electrons in an s band coupled by s-f exchange to a lattice of localized spins. The approximation used was the RPA, which means that in the limit of a vanishing s-f coupling constant the dielectric function reduces to the standard self-consistent-field expression[17] and the susceptibility reduces to the expression of Kawasaki and Mori.[21] The main weakness of our calculation is the neglect of spin-flip terms in the s-f exchange Hamiltonian. Their effect can be studied in detail at low temperatures using the spin-wave approximation. The linear response coefficients are then found to be of the general form of our results, Eqs. (27)–(32), provided one reinterpretes the s-f coupling constant I as well as the electronic and magnetic susceptibilities, $\chi_{0\sigma}$ and χ^{H}_{zz}, to be renormalized. Consequently, the value of the quantity j will no longer be given by Eq. (55). We have not been able to determine the renormalized terms quantitatively. Therefore, there is no convincing argument to reject the value of j obtained by Eq. (55) in favor of one that is substantially smaller. The work of Nagaev and Grigin,[12] furthermore, suggests that the coupling constant j might even be dependent on temperature and carrier density. At any rate, since it is the product of the quantity j with the electronic and magnetic susceptibilities of the two subsystems that appears as a measure of the coupling strength between spin- and charge-density fluctuations, it is plausible that the response obtained without spin flips between the two subsystems is too strong. Our reduction of j is hoped to correct for this in a phenomenological way and avoids instabilities in the response coefficients of the translationally invariant system.

Using the RPA results of Sec. II, we attempted in Sec. III to describe the IMT in Eu-rich EuO. We calculated the instability of the metallic phase toward formation of BMP[9] using a variational technique. In particular, the doubly charged oxygen vacancies in Eu-rich EuO were assumed to trap one electron each in a tightly bound state, the other electron being in the conduction band in the metallic phase. At first we neglected the effect of the tightly bound electron, except for a cancellation of one vacancy charge. The critical carrier density was then computed as a function of temperature by varying the binding energy of a conduction electron in the field of the vacancy screened by the dielectric function. In addition, the vacancy polarizes the spins of the Eu^{++} ions in a region around the vacancy where the electron is about to become localized. This and the dependence of the dielectric response on the long-range magnetic order make the critical carrier density temperature dependent. This is qualitatively in accord with the calculation of Leroux-Hugon.[13] But since Leroux-Hugon (besides using different numerical parameters, an Ornstein-Zernike expression for the longitudinal susceptibility, and the Thomas-Fermi approximation) neglected the spin-down conduction band, which will be populated close to the Curie temperature T_C, his results do not show a strong decrease of the critical carrier density at T_C. Based on the simple Mott criterion we showed, however, that this decrease is an essential property of calculations of this type, but it is not in accord with experimental results.

One could argue that short-range correlations of the magnetization close to T_C will smear out the decrease at T_C (cf. Fig. 1) and shift it to higher temperatures. However, one can roughly estimate the correlation length with the Ornstein-Zernike relation and compare it with the crystal-Bohr radius. It is plausible then that short-range correlations will have no effect on the binding energy once the correlation length is well below the crystal-Bohr radius. This way we obtain an estimate of the shift to be of the order of 1 K. This is still much too close to T_C, especially in view of the peak in the resistivity above T_C.[10] It is also unlikely that self-trapping of the charge carriers of the type considered by Yanase will alter the decrease of n_c near T_C substantially since it is estimated not to occur in the Eu chalcogenides.[28]

When the tightly bound impurity electron is not neglected, it can be shown to give rise to a deviation from the Weiss molecular field. This deviation of the molecular field is believed to dominate self-trapping effects since it is strongly localized at the impurity. It is found to increase the critical carrier density for the instability of the metallic state towards formation of a triplet impurity state, even well above T_C (Fig. 2). Since local spin deviations are mainly responsible for increased electron binding of the impurity, the localizing effect is strongest near T_C where the magnetic susceptibility is large and large spin deviations from the mean value are easily realized. The instability of the metallic state toward formation of a triplet impurity state does not preclude that the actual ground state of the heliumlike impurity is a singlet in the nonmetallic state, since our calculation can only give the *instability* of the metallic phase.

Our results allow a range of the carrier density for which the IMT occurs below T_C and activated behavior persists well into the paramagnetic regime as is observed in same samples. Also it should be noted that the density range over which the IMT can occur is substantially wider than that indicated in Fig. 1 or predicted by previous calculations.[13] Although the curves of Fig. 2 give qualitative agreement with experiment, the prediction that the IMT occurs near 30 K is not in agreement with the observed transition temperature of about 50 K. It also appears that samples which are metallic at very low temperatures are predicted to undergo a second transition to a metallic state at about 100 K. This transition is not observed experimentally. If, however, the exchange parameter $\tilde{J}$ [Eq. (51)] is chosen even larger than we did and/or if the spatial extend of the tightly bound electron is increased, this second transition temperature can be increased easily to much higher temperatures where the conduction electrons cease to be degenerate (one can estimate the Fermi temperature of the samples under consideration to be about 400 K). Furthermore, Fig. 2 indicates that for carrier densities n below 3×10^{19} cm^{-3} the IMT temperature should decrease rapidly with decreasing n, a fact which has not been experimentally observed.[10] One reason for this could be our choice of $m^*=0.55m_0$. Decreasing m^* further would lower the set of curves in Fig. 2, thereby increasing the IMT temperature, but an m^* of $0.55m_0$ seems already rather small.[6,33] Inaccuracies in our description—besides the temperature- and carrier-density independent coupling constant j—may also stem from the use of Eq. (40) for the longitudinal susceptibility. Although it is clear that a correct *longitudinal* susceptibility should be used below T_C, it is not clear that Eq. (40) serves this purpose best, partly because it is a long-wavelength approximation and partly because it is not too well established experimentally.[34]

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