Magnetic and nonmagnetic impurities in two-dimensional metals

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We examine the spatial dependence of the spin polarization arising from a magnetic impurity as well as the impurity-impurity interaction for impurities dissolved in an idealized two-dimensional metal. We find that the usual $(x \cos x - \sin x)/x^4$ dependence of the oscillations in a three-dimensional metal is replaced by a $\sin x/x^2$ dependence in a two-dimensional system, where $x = 2k_F r$, k_F is the Fermi wave vector, and r is the distance from the impurity. Using this result we show that the temperature and concentration dependence of the thermodynamic properties of a magnetic-impurity system dissolved in a two-dimensional metal will, in the molecular-field approximation, be identical with that dissolved in a three-dimensional system. Similarly, the distance dependence of the screening charge from a nonmagnetic impurity in a metal changes from a $1/r^3$ (times an oscillating function) dependence in three dimensions to a $1/r^2$ dependence in two dimensions.

I. INTRODUCTION

There has been a great deal of interest in the properties of magnetic impurities dissolved in nonmagnetic metal hosts. The earlier workers 1^{-5} addressed themselves to the behavior of the conduction-electron-spin polarization and the impurityimpurity interaction potential, and found that the spatial dependence of the spin polarization (impurity-impurity interaction potential) is proportional to $(x \cos x - \sin x)/x^4$, where $x = 2k_F r$, k_F is the Fermi wave vector and r is the distance from the impurity (between the two impurities).

The behavior of the charge density around a nonmagnetic impurity in metal hosts has also been treated by several investigators.^{6,7} As in the magnetic-impurity case, it was found that the screening charge oscillates in sign as a function of position r and decreases as r^{-3} for large r.

The works mentioned above treated an impurity system dissolved in three-dimensional metals. However, as far as we know, no treatment exists on the properties of magnetic impurities dissolved in two-dimensional metals. The theoretical predictions of such a treatment may be of interest in view of the recent progress made in performing measurements on two-dimensional systems.⁸ Such measurements could possibly shed some light on the interaction mechanism in the two-as well as three-dimensional alloy system as will be discussed later.

The purpose of this paper is to examine the behavior of magnetic impurities dissolved in an idealized two-dimensional system using perturbation theory. We find that the behavior of the spin polarization around a magnetic impurity as well as the range of the impurity-impurity interaction changes from $1/r^3$ dependence times an oscillating function in three dimensions to a $1/r^2$ dependence

in a two-dimensional metal host. A similar result holds for the behavior of the screening charge caused by a nonmagnetic impurity.^{7,9}

Using these results for a set of magnetic impurities randomly distributed in a nonmagnetic metal host, we show that in the mean random molecular-field approximation (MRFA),¹⁰ the concentration and temperature dependence of the thermodynamic properties of the system will be identical in two and three dimensions. The predicted behavior of the thermodynamic properties for impurities dissolved in an ideal two-dimensional system can be summarized as follows: (a) The low-temperature specific heat should be proportional to the temperature T and be independent of the impurity concentration c. (b) The magnetic susceptibility χ is predicted to have a broad maximum, with the temperature of the maximum proportional to the impurity concentration. Similarly, near $T \rightarrow 0$, χ is predicted to be independent of c. For further references of the thermodynamic behavior of the system the reader is referred to several works on the subject.¹⁰⁻¹⁴

II. CONDUCTION-ELECTRON POLARIZATION FROM A LOCALIZED MAGNETIC IMPURITY

Starting from the many-impurity s-d Hamiltonian used by Kasuya,² Yosida,³ and Mitchell⁵ we assume a Hamiltonian ${\mathcal K}$ of the form

$$\mathcal{F} = \mathcal{F}_{0} + \mathcal{F}_{s-d} , \qquad (2.1)$$

where

$$\mathcal{K}_{0} = \sum_{k\sigma} \epsilon_{k\sigma} c_{k\sigma}^{\dagger} c_{k\sigma}$$

and

$$\mathcal{H}_{s-d} = \frac{J}{2N} \sum_{k'} \sum_{i} (\bar{\sigma}_{\alpha\beta} \cdot \bar{\mathbf{S}}_{i}) e^{i(\vec{\mathbf{k}} - \vec{\mathbf{k}}') \cdot \vec{\mathbf{R}}_{i}} c_{k\alpha}^{\dagger} c_{k'\beta} .$$
(2.2)

11

 $\sigma_{\alpha\beta}$ is the expectation value of the Pauli spin matrix between the spin states $|\alpha\rangle$ and $|\beta\rangle$, $c_{k\alpha}^{\dagger}$ and $c_{k\beta}$ are the respective creation and annihilation operators for an electron with wave vector k and spin σ , S_i is the impurity spin located at position R_i , J is the *s*-*d* exchange interaction assumed to be a constant, and N is the number of the sites in the solid. The summation over *i* indicates a summation over all impurities.

From Eq. (2.2) we obtain an effective impurityimpurity interaction Hamiltonian \mathcal{W}_{ij} for two impurities i and j of the form

$$\mathcal{H}_{ij} = \left(\frac{J}{N}\right)^2 \left(\vec{\mathbf{S}}_i \cdot \vec{\mathbf{S}}_j\right) \sum_{\substack{k \le k_F \\ k' > k_F}} \frac{e^{i\left(\vec{\mathbf{k}} - \vec{\mathbf{k}}'\right) \cdot \left(\vec{\mathbf{R}}_i - \vec{\mathbf{R}}_j\right)}}{\epsilon_k - \epsilon_{k'}} \quad . \tag{2.3}$$

A similar expression was obtained by Yosida³ for the conduction-electron polarization density $\Delta \rho = \rho_+ - \rho_-$, where ρ_+ (ρ_-) is the density for spin up (spin down):

$$\Delta \rho \propto \left(\frac{2J}{N}\right) S_n^z \sum_{\substack{k \leq k_F \\ k' > k_F}} \frac{e^{i\left(\vec{k} - \vec{k}\,'\right) \cdot \vec{t}}}{\epsilon_k - \epsilon_{k'}} \quad , \tag{2.4}$$

where S_n^r is the *z* component of spin \hat{S} , *V* is the volume of the solid, and *r* is the distance from the impurity.

We next derive the expression for the conductionelectron polarization and the impurity-impurity interaction potential for a two-dimensional metal. Let $\vec{k} - \vec{k}' = \vec{q}$, $\vec{R}_i - \vec{R}_j = \vec{r}_{ij}$ and let the summation in Eq. (2.3) be denoted by *I*, then changing the summation in Eqs. (2.3) and (2.4) to an integration in two dimensions and letting $\epsilon_k = \hbar^2 k^2/2 m^*$, where m^* is the effective mass of the electron, we obtain

$$I = -\frac{2m^*}{\hbar^2} \frac{V}{(2\pi)^2} \sum_{q \ge 0} \frac{e^{i \frac{q}{q} \cdot \frac{r}{T}}}{q}$$
$$\times \int_0^{k_F} k \, dk \int_0^{2\pi} \frac{d\theta}{q - 2k \cos\theta} \quad . \tag{2.5}$$

Evaluating the integral for the principal part of $(q - 2k\cos\theta)^{-1}$ gives

$$\int_{0}^{2\pi} \frac{d\theta}{q - 2k \cos \theta} = \begin{cases} \frac{2\pi}{[q^2 - (2k)^2]^{1/2}} & , q > 2k \\ 0 & , q < 2k \end{cases}$$
(2.6)

Using Eq. (2.6) in Eq. (2.5) and integrating over the angles between the vectors \vec{q} and \vec{r} gives

$$I = -\frac{V^2}{(2\pi)^2} \frac{2m^*}{\hbar^2} \int_0^{k_F} k \, dk \int_{2k}^{\infty} dq \, J_0(qr) \frac{1}{[q^2 - (2k)^2]^{1/2}}$$
(2.7)

In Eq. (2.7) q is restricted to values such that q > 2k. To obtain Eq. (2.7) we used the relationship

$$\frac{1}{2\pi} \int_0^{2\pi} e^{ix \cos y} \, dy = J_0(x) \,,$$

where J_l is the Bessel function of order l. Integrating Eq. (2.7) first over q, and using the relation¹⁵

$$\int_{1}^{\infty} dy \, \frac{J_0(2kry)}{(y^2-1)^{1/2}} = \frac{1}{2}\pi N_0(kr) J_0(kr) \,, \qquad (2.8)$$

Eq. (2.7) becomes

$$I = -\frac{m^* V^2}{4\pi \hbar^2} \frac{1}{r^2} \int_0^{x_F} x \, dx \, J_0(x) \, N_0(x)$$
$$= -\frac{m^* V^2}{4\pi^2 \hbar^2} \frac{x_F^2}{4r^2} [2J_0(x_F)N_0(x_F) + 2J_1(x_F)N_1(x_F)] ,$$
(2.9)

where $x_F = k_F r$ and N_l is the Neuman function of order *l*. The integral, Eq. (2.9), was obtained using the relationship¹⁶ valid for Bessel and Neuman functions of integer order only.

For the case when $2k_F r \gg 1$, we use the first two terms of the asymptotic form of the Bessel and Neuman functions¹⁵ to obtain

$$I = \frac{V^2}{16\pi^2} \frac{2m^*}{\hbar^2} \left[\frac{\sin 2k_F r}{r^2} - \frac{13}{16k_F} \frac{\cos 2k_F r}{r^3} + O\left(\frac{1}{r^4}\right) \right].$$
(2.10)

We remark that in this paper we address ourselves to dilute concentrations of impurities dissolved in two-dimensional metals, such that $2k_F r_{av} >> 1$, where r_{av} is the average interimpurity distance. Thus Eq. (2.10) can be used for layered transition-metal dichalcogenides^{16a} as well as the twodimensional semiconductor systems.⁸ Substituting Eq. (2.10) into Eq. (2.9) we obtain for the spin polarization of the conduction electrons (for large r)

$$\Delta \rho(r) \propto \frac{n}{4\pi} \left(\frac{J}{E_F} \right) S_{\pi}^{z} \left[\frac{\sin(2k_F r)}{r^2} - \frac{13}{16k_F} \frac{\cos 2k_F r}{r^3} + O\left(\frac{1}{r^4}\right) \right],$$
(2.11)

where *n* is the number of conduction electrons per lattice site. Similarly, substituting Eq. (2.10) into Eq. (2.3) we obtain an effective impurity-impurity interaction Hamiltonian \Re_{ij} of the form

$$\begin{aligned} \mathfrak{K}_{ij} &= -\frac{n^2}{4} \left(\frac{J^2}{E_F} \right) \left(\mathbf{\tilde{S}}_i \cdot \mathbf{\tilde{S}}_j \right) \\ &\times \left[\frac{\sin(2k_F r)}{(k_F r)^2} - \frac{13}{16} \frac{\cos 2k_F r}{(k_F r)^3} + O\left(\frac{1}{r^4} \right) \right] \\ &\equiv v_{ij} \left(\mathbf{\tilde{S}}_i \cdot \mathbf{\tilde{S}}_j \right) . \end{aligned}$$
(2.12)

It is interesting to note that the $(\cos 2k_F r)/r^3$ dependence of the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction in three-dimensional systems is replaced by a $(\sin 2k_F r)/r^2$ dependence in two dimensions. As will be seen in Sec. III this result has important consequences with regard to the scaling of the thermodynamic properties of the dilute magnetic alloys in two and three dimensions.

III. SCALING OF THE THERMODYNAMIC PROPERTIES IN TWO AND THREE DIMENSIONS

The thermodynamic properties of a set of randomly distributed magnetic impurities in three dimensions has been considered by several authors in the molecular-field approximation.^{10, 17, 18} In this section we show that in the molecular-field approximation the concentration and temperature dependence of thermodynamic properties for the two- and three-dimensional random impurity system will be identical.

To show this, we define the effective field H_0 at an arbitrary impurity site 0, by the relation

$$H_0 = \sum_j v_{0j} \langle \mu_j \rangle , \qquad (3.1)$$

where v_{0j} is given by Eq. (2.12) and $\langle \mu_j \rangle$ is the thermodynamic average of the Ising spin at site j, which experiences an effective field H_{j} .²⁰ Since the magnetic impurities are randomly distributed in the system, r_{0j} and hence v_{0j} and H_j are random variables. We wish to find the self-consistent probability distribution of H_0 . This can be done using the formalism developed by one of the authors.¹⁰ We denote this method as the meanrandom-field (MRF) approximation. To find the formal expression for the probability distribution we use the statistical model of Margenau,¹⁹ to obtain

$$P(H) = \sum_{v_n} \int_{r_n} \delta\left(H - \sum v_{0j} \langle \mu_j \rangle\right)$$
$$\times P(\mathbf{\bar{r}}_{01}, \cdots, \mathbf{\bar{r}}_{0n}) d^2 \mathbf{\bar{r}}_{0n} , \qquad (3.2)$$

where $P(\vec{r}_{01}, \dots, \vec{r}_{0n}) d^2 r_{0n}$ is the probability of finding particle 1 in a volume $d^2 r_1$, and particle *n* in a volume $d^2 r_n$. The symbol v_n under summation indicates a sum over each potential having the value of $\pm a/r_{0j}^3$ with probability $\frac{1}{2}$ as is discussed in Ref. 10.

Going through the steps outlined in Ref. 10, we obtain similar equations as in Ref. 10 except that V' in Eq. (2.15) becomes in two dimensions

$$V' = \int_{-\infty}^{\infty} P(H) dH \int_{0}^{\infty} \left[1 - \cos\left(\frac{\rho a \langle \mu \rangle}{r^{2}}\right) \right] r dr.$$
(3.3)

Changing variables by letting $z = |a\rho\langle\mu\rangle| / r^2$ we obtain

$$V' = \pi |\rho a| \| \mu \| \int_0^\infty \frac{1 - \cos z}{z^2} dz , \qquad (3.4)$$

where $\|\mu\|$ is the magnitude of the thermal average of the spin averaged over all allowable internal fields, i.e.,

$$\| \mu \| = \int_{-\infty}^{\infty} P(H) |\tanh\beta H| \, dH \, . \tag{3.5}$$

Thus all equations and relations of Ref. 10 hold in two dimensions except that the width of the distribution function in two dimensions, Δ_2 , is modified from its value in three dimensions, Δ_3 ,

$$\Delta_2 = \frac{3}{4} \Delta_3 = \frac{1}{2} \pi^2 |a| n_{0n} c ||\mu|| , \qquad (3.6)$$

where a is the strength of the interaction at a distance of one lattice constant, Δ_n denotes the *n*-dimensional width of the distribution function, n_{on} is the number of sites per unity cell in *n* dimensions, and c is the fractional impurity concentration. The probability distribution for the field H in two as well as in three dimensions is thus in the Ising model

$$P(H, T, c) = \frac{1}{\pi} \frac{\Delta_n}{\Delta_n^2 + H^2} \quad . \tag{3.7}$$

Equation (3.7) is an integral equation for the width of the probability distribution Δ_n . This can be seen by substituting Eqs. (3.5) and (3.7) into Eq. (3.6). We thus get

$$\Delta_n(\beta) = \frac{2}{\pi} \int_0^\infty \frac{\Delta_n(\beta)}{[\Delta_n(\beta)]^2 + H^2} |\tanh\beta H| \, dH \, .$$
(3.8)

Equation (3.8) gives a self-consistent solution for $\Delta_n(\beta)$ for all β , and allows one to obtain the temperature dependence of the system for all *T*. All the thermodynamic properties of the system are obtained (in the approximation used) as follows:

$$y(\beta, c) = N_0 c \int_{-\infty}^{\infty} y(H, \beta) P(H, \beta, c) dH$$
, (3.9)

where N_0 is the number of sites in the solid, c is the impurity concentration, $y(H, \beta)$ is the single impurity thermodynamic function in a fixed field H (y may be the magnetization M, or the magnetic susceptibility χ , or the magnetic specific heat $C_{\rm M}$) and temperature $(k_{\rm B}\beta)^{-1}$, where $k_{\rm B}$ is the Boltzmann constant. The consequence of the last two equations is that since the two- and threedimensional systems have similar field distributions, all the concentration and temperature dependence of the thermodynamic properties of the magnetic impurity system will be the same in two and three dimensions.²¹ Thus measurements on a two-dimensional system should give: (a) The low-temperature specific heat is proportional to the temperature T and independent of the impurity concentration c. (b) The magnetic susceptibility $\chi(T)$ for low temperature is $\chi(T, c)$ =A + B(T/c), where A and B are independent of c and T. We also predict that $\chi(T)$ has a maximum as a function of temperature. (The possibility of observing cusps in the magnetic susceptibility similar to that in a three-dimensional alloy systems should not be excluded.) The high-temperature properties should also be equivalent to those obtained for the three-dimensional system.

One can further argue that should such an "idealized" two-dimensional system be found say in one of the layered two-dimensional metallic compounds, some important questions in connection with the alloy problem could be studied and possibly settled. In order to clarify this point we return briefly to the present understanding of the three-dimensional magnetic alloy system.

The current understanding of the very-low-temperature properties of these alloys is based on the (random) molecular-field approximation and the Ising model.^{10,17} The validity of the Ising model has been questioned by several investigators^{22,23} and it was argued that a realistic Heisenberg-model treatment should give a T^3 dependence of the magnetic specific heat C_M rather than the experimentally measured linear dependence with temperature. Let us assume for the moment that the part of C_M arising from the molecular-field approximation should really be treated using a Heisenberg model and should really give a T^3 dependence to C_M . Then we must argue that the linear dependence in C_M is due to some other mechanism (such as spin waves, or spin-spin correlations) which is not understood at the moment. Any spin-wave contribution to C_{M} is expected to differ sharply in two and three dimensions. Therefore, measuring C_M for a magnetic impurities in a two-dimensional (metallic) layer compound can shed further light on the nature of the specific-heat contribution of the magnetic impurities at low temperatures. In particular, should C_M be independent of the dimensionality of the system, this would indicate that C_M arises from the Ising-like molecular-field approximation, still leaving the question open of why the Ising

model is valid. It is thus seen that an interplay between the behavior of the two- and three-dimensional systems may clarify the understanding of both.

IV. CHARGED IMPURITIES

The static dielectric function of the two-dimensional metal is $\epsilon(q) = 1 + 4\pi \chi(q)$, where $\chi(q)$ is the Lindhard-like expression²⁴ for two dimensions:

$$\chi(q) = \frac{e^2}{2Vq} \sum_{k} \frac{f^0(\epsilon_k) - f^0(\epsilon_{k+q})}{\epsilon_k - \epsilon_{k+q}} , \qquad (4.1)$$

where f^0 is the zero-temperature Fermi function approximated by its unperturbed value and V is the two-dimensional volume of the solid. By calculating the sum in Eq. (4.1) in two dimensions, we obtain for the dielectric function $\epsilon(q)$:

$$\epsilon(q) = \begin{cases} 1 + \frac{\alpha}{q} - \frac{\alpha}{q} \left[1 - (2k_F)^2/q^2 \right]^{1/2}, & q \ge 2k_F \\ \\ 1 + \frac{\alpha}{q} & q < 2k_F \end{cases},$$
(4.2)

where $\alpha = e^2 m^*/\hbar^2$. The $\epsilon(q)$ is not to be confused with the value of ϵ used previously for the energy. The effective potential v(r) in the metal becomes

$$v(r) = \sum_{q} \frac{v_{q}^{0}}{\epsilon(q)} e^{i\vec{q}\cdot\vec{r}}, \qquad (4.3)$$

where v_q^0 is the *q* component of the unperturbed potential. Replacing the summation by an integral in two dimensions gives

$$v(r) = \frac{V}{2\pi} \int_0^\infty q \, dq \, J_0(qr) v_q^0 / \epsilon(q) \,, \qquad (4.4)$$

where $J_1(x)$ is the Bessel function of order *l*. Integrating Eq. (4.4) by parts, assuming that v_q^0 goes to zero for $q \to \infty$ faster than $q^{-1/2}$, we obtain

$$v(r) = \frac{V}{2\pi r} \int_0^\infty q \ J_1(qr) \frac{d}{dq} \left(\frac{v_q^0}{\epsilon(q)}\right) dq \ . \tag{4.5}$$

Equation (4.5) is evaluated for large r using the method outlined by Harrison.²⁴ Then we have

 $= (\pi a/k_F r) J_{1/2}(k_F r) N_{1/2}(k_F r)$,

$$v(r) = (-2a/r) \int_{2k_F}^{\infty} \frac{J_1(qr)dq}{[q^2 - (2k_F)^2]^{1/2}}$$

(4.6)

where

$$a = V \alpha (2k_F) v^0 (2k_F) / 16 \pi \epsilon^2 (2k_F)$$
,

 $v^{0}(2k_{F})$ and $\epsilon(2k_{F})$ are the values of the unscreened potential and dielectric function at $2k_{F}$, and α has been defined immediately following Eq. (4.2). Substituting the values of $J_{1/2}(x)$, and $N_{1/2}(x)$, we obtain <u>11</u>

 $v(r) = a (\sin 2k_F r) / (k_F r)^2$. (4.7)

This result was found previously by Stern⁹ using a different approach.

It is interesting to note that, just like in the magnetic impurity case, the r^{-k} dependence of the interaction in k dimensions (k=2, 3) causes

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some similarities in the screening properties of two- and three-dimensional alloys. The modification of the potential for two-dimensional metals will have important consequences in interpreting NMR experiments in such alloys, using similar arguments as was done by Rowland^{25,26} and Kohn and Vosko⁷ for three-dimensional metals.

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