heat pulses in the [100] direction in pure LiF, Si, and Ge is also qualitatively accounted for by the above theory. For LiF the ratio of the elastic constants $C_{11}:C_{12}:C_{44}$ at low temperatures is 1:0.34:0.52, whereas for Si and Ge these ratios are 1:0.39:0.48 and 1:0.38:0.52, respectively. The deviation between the group and phase velocity does not depend on the magnitude of the elastic constants, but only on their ratios. Since these ratios are very similar for LiF, Si, and Ge, one expects similar heat-pulse propagation in these solids. In contrast, the ratio for KCl is 1:0.11: 0.14. Since for NaF the ratio is 1:0.21:0.27, its heat-pulse propagation properties are expected to be intermediate between those of LiF and KCl, which was indeed observed.7

The experimental results are consistent with the above theory; phonon focusing in solids has thus been demonstrated. An investigation of heatpulse propagation in other directions in LiF and KCl is currently being conducted.

The authors thank Mr. T. Kubaska and Mr. T. Fjeldly for obtaining heat-pulse data in KCl. *Research supported in part by the National Science Foundation.

 1 R. J. von Gutfeld and A. H. Nethercot, Jr., Phys. Rev. Letters <u>12</u>, 641 (1964).

²R. J. von Gutfeld and A. H. Nethercot, Jr., in <u>Proceedings of the Ninth International Conference on Low</u> <u>Temperature Physics, Columbus, Ohio, 1964</u>, edited by J. G. Daunt (Plenum Press, New York, 1965), p. 1189.

³R. J. von Gutfeld and A. H. Nethercot, Jr., Phys. Rev. Letters 17, 868 (1966).

⁴J. M. Andrews and M. W. P. Strandberg, Proc. IEEE <u>54</u>, 523 (1966).

⁵R. J. von Gutfeld, <u>Physical Acoustics</u>, edited by W. P. Mason (Academic Press, Inc., New York, 1968), Vol. 5, p. 233.

⁶M. Pomerantz and R. J. von Gutfeld, in <u>Proceedings</u> of the International Conference on the Physics of Semiconductors, Moscow, U. S. S. R., 1968 (Nauka Publishing House, Leningrad, U.S.S.R., 1968), Vol. 2, p. 690. ⁷S. J. Rogers and R. J. Rollefson, Bull. Am. Phys.

Soc. <u>12</u>, 339 (1967).

⁸M. J. P. Musgrave, Proc. Roy. Soc. (London), Ser. A <u>226</u>, 339 (1954).

⁹G. F. Miller and M. J. P. Musgrave, Proc. Roy. Soc. (London), Ser. A <u>236</u>, 352 (1956).

CHARGE AND SPIN SUSCEPTIBILITY OF A FERROMAGNETIC ELECTRON GAS

D. J. Kim, H. C. Praddaude, and Brian B. Schwartz Francis Bitter National Magnet Laboratory,† Massachusetts Institute of Technology, Cambridge, Massachusetts 02139 (Received 26 June 1969)

We have obtained the charge and spin susceptibilities in the ferromagnetic state of an electron gas, including the effects of the long-range nature of the Coulomb interaction from a unified point of view. The susceptibilities are discussed as a function of the magnetization for typical ferromagnetic alloy systems.

In this Letter we present a general formulation for the charge and spin responses of a ferromagnetic electron gas to a charge potential or magnetic field. The long-range effects of the Coulomb interaction are adequately accounted for within the random-phase approximation while insuring charge conservation. Besides obtaining the charge susceptibility to a charge potential, $\chi_{e\,e}$, and the spin susceptibility to a magnetic field, χ_{mm} , we also derive two additional nondiagonal susceptibilities, i.e., the spin response to a charge potential χ_{me} and the charge response to a magnetic field χ_{em} . The nondiagonal susceptibilities do not appear in the paramagnetic state and have not been previously treated from a general viewpoint. To interpret the electron spin polarization and magnetic moment produced by impurities in ferromagnets Friedel¹ and others² have presented a physical picture for the spin imbalance associated with the screening charge about an impurity in a ferromagnetic metal which in a sense corresponds to χ_{me} . Since the nondiagonal susceptibility we derive can be as important as the usual diagonal susceptibility, both must be included in any analysis of the total charge or spin responses of ferromagnets.

We use the following Hamiltonian to calculate the spin and charge response functions for an interacting electron gas:

$$\mathcal{H} = \sum_{I,\sigma} \epsilon_{I\sigma} c_{I\sigma}^{\dagger} c_{I\sigma} + \frac{1}{2} \sum_{\substack{I,I',\kappa\\\sigma,\sigma'}} V(\kappa) c_{I\sigma}^{\dagger} c_{I'\sigma'}^{\dagger} c_{I'-\kappa,\sigma'} c_{I+\kappa,\sigma} + \mathcal{H}_{\alpha'} \quad (\alpha = m \text{ or } e),$$
(1)

419

where the magnetic field and charge potential perturbations are given by

$$\mathscr{K}_{m'} = \mu_{B} H(q) \sum_{I} (c_{I+} {}^{\dagger} c_{I-q,+} - c_{I-} {}^{\dagger} c_{I-q,-}), \qquad \mathscr{K}_{e'} = e V_{0}(q) \sum_{I} (c_{I+} {}^{\dagger} c_{I-q,+} + c_{I-} {}^{\dagger} c_{I-q,-}).$$
(2a, 2b)

The first term of Eq. (1) is the kinetic energy of the conduction electrons and $c_{I\sigma}^{\dagger}$ is the creation operator of a conduction electron with energy ϵ_I and spin σ (= + or -). The second term is the Coulomb repulsion between electrons and $V(\kappa) = 4\pi e^2/\kappa^2$. The prime on the summation means to exclude $\kappa = 0$. H(q) and $V_0(q)$ are the external magnetic field and charge potential with wave number q, μ_B is the Bohr magneton, and e is the electron charge.

Four susceptibilities can be defined as follows: the spin and charge susceptibility to a magnetic field, respectively,

$$\chi_{mm}(q) = \frac{\mu_{\rm B}[n_+^{m}(q) - n_-^{m}(q)]}{H(q)}, \quad \chi_{em}(q) = \frac{e[n_+^{m}(q) + n_-^{m}(q)]}{H(q)};$$
(3a, 3b)

and the spin and charge susceptibility to a charge potential given by

$$\chi_{me}(q) = \frac{\mu_{B}[n_{+}^{e}(q) - n_{-}^{e}(q)]}{-V_{0}(q)}, \quad \chi_{ee}(q) = \frac{e[n_{+}^{e}(q) + n_{-}^{e}(q)]}{-V_{0}(q)}, \quad (3c, 3d)$$

where $n_{\sigma}^{\alpha}(q) = \sum_{k} \langle c_{k\sigma}^{\dagger} c_{k+q,\sigma} \rangle$ with $\alpha = m$ or e for the magnetic or charge perturbation \mathcal{H}_{m}' or \mathcal{H}_{e}' , the angular brackets indicate the thermal average, and the z axis is taken in the direction of the magnetization of the system.

Using the two-time Green's function³ $\langle c_{k+q,\sigma} | c_{k\sigma}^{\dagger} \rangle_{\omega}$, $n_{\pm}(q)$ is calculated by the well-known procedure from

$$\langle a^{\dagger}b\rangle = -\frac{1}{2\pi} \int_{-\infty}^{\infty} d\omega \{\langle b | a^{\dagger} \rangle_{\omega - i0^{+}} - \langle b | a^{\dagger} \rangle_{\omega - i0^{+}} \} f(\omega), \tag{4}$$

where $f(\omega)$ is the Fermi function and the equation of motion for $\langle c_{k+q,q} | c_{kq}^{\dagger} \rangle_{\omega}$ is

$$(\omega - \epsilon_{k+q}) \langle c_{k+q,\sigma} | c_{k\sigma}^{\dagger} \rangle_{\omega} = \delta_{q,0} + W_{\sigma}^{\alpha}(q) \langle c_{k\sigma} | c_{k\sigma}^{\dagger} \rangle_{\omega} + \sum_{I,\kappa,\sigma'} V(\kappa) \langle c_{I\sigma'}^{\dagger} c_{I-\kappa,\sigma'} c_{k+q+\kappa,\sigma} | c_{k\sigma}^{\dagger} \rangle_{\omega},$$
(5)

where $W_{\pm}^{m}(q) = \mp \mu_{B}H(q)$ and $W_{\pm}^{e}(q) = eV_{0}(q)$. The problem here is how to handle the last term on the right-hand side of Eq. (5). We simplify this Green's function using the random-phase approximation:

$$\sum_{I,\kappa,\sigma'} V(\kappa) \langle c_{I\sigma'}{}^{\dagger} c_{J-\kappa,\sigma'} c_{k+q+\kappa,\sigma} | c_{k\sigma}{}^{\dagger} \rangle_{\omega} = V(q) [n_{+}(q) + n_{-}(q)] \langle c_{k\sigma} | c_{k\sigma}{}^{\dagger} \rangle_{\omega} (1 - \delta_{q,0})$$
$$- \sum_{\kappa} ' V(\kappa) \langle c_{k+q+\kappa,\sigma}{}^{\dagger} c_{k+q+\kappa,\sigma} \rangle \langle c_{k+q,\sigma} | c_{k\sigma}{}^{\dagger} \rangle_{\omega}$$
$$- \sum_{\kappa} ' V(\kappa) \langle c_{k+\kappa,\sigma}{}^{\dagger} c_{k+q+\kappa,\sigma} \rangle \langle c_{k\sigma} | c_{k\sigma}{}^{\dagger} \rangle_{\omega} (1 - \delta_{q,0}).$$
(6)

The physical meaning of each term on the right-hand side of Eq. (6) is very clear if we substitute Eq. (6) into Eq. (5). The first term on the right-hand side of Eq. (6) represents the effect of the Coulomb potential due to charge polarization $[n_+(q) + n_-(q)]$. Accordingly V(q) appearing here should be the bare Coulomb interaction. The second term is the exchange self-energy which gives rise to the spin splitting of the band. This term may be incorporated to ϵ_{k+q} on the left-hand side of Eq. (5) changing ϵ_{k+q} to ϵ_{k+qo} . The last term in Eq. (6) is responsible for the exchange enhancement of the magnetic susceptibility. In the paramagnetic case a higher order treatment of the Green's-function equation results in a screening of the potential appearing in the last two terms.⁴ Although the explicit expression for the screened exchange potential is complicated, as was shown for the paramagnetic case,⁴ we follow a procedure similar to Hubbard⁵ where we replace the summation over κ in the last term of Eq. (6) by $U(q)n_o(q)$, where U(q) is an effective exchange interaction. The procedure is also supported by the physical reasoning that the exchange field which gives rise to the exchange enhancement should be of the form of an effective exchange interaction times the spin density $-U(q)[n_+(q)-n_-(q)]$. Similarly in the second term of Eq. (6), $V(\kappa)$ is replaced by U(0) since the zero-wave-number exchange field is responsible for the spin splitting of the band. Notice that the spin susceptibility derived by Izuyama, Kim, and Kubo⁶ (IKK) would be obtained if we replace $V(\kappa)$ by U in all three terms in Eq. (6). The final approximation in Eq. (6) is to put $\langle c_{k\sigma} | c_{k\sigma}^{\dagger} \rangle_{\omega} \sim (\omega - \epsilon_{k\sigma} + i0^{\dagger})^{-1}$. Using these approximations we

(3d')

obtain

2

$$n_{\sigma}^{\alpha}(q) = -W_{\sigma}^{\alpha}(q)F_{\sigma}(q) - V(q)[n_{+}^{\alpha}(q) + n_{-}^{\alpha}(q)]F_{\sigma}(q) + U(q)n_{\sigma}^{\alpha}(q)F_{\sigma}(q),$$
⁽⁷⁾

where $F_{\sigma}(q) = -\sum_{k} [f(\epsilon_{k\sigma}) - f(\epsilon_{k+q,\sigma})](\epsilon_{k\sigma} - \epsilon_{k+q,\sigma})^{-1}$. Solving the coupled Eq. (7) for n_{σ}^{α} we obtain the spin and charge susceptibilities:

$$\chi_{mm}(q) = \mu_{\rm B}^2 \frac{\tilde{F}_+(q) + \tilde{F}_-(q) + 4V(q)\tilde{F}_+(q)\tilde{F}_-(q)}{1 + V(q)[\tilde{F}_+(q) + \tilde{F}_-(q)]},\tag{3a'}$$

$$\chi_{em}(q) = \chi_{me}(q) = \mu_{\rm B} e \frac{\tilde{F}_{+}(q) - \tilde{F}_{-}(q)}{1 + V(q) [\tilde{F}_{+}(q) + \tilde{F}_{-}(q)]}, \tag{3b', 3c'}$$

$$\chi_{ee}(q) = e^2 \frac{F_+(q) + F_-(q)}{1 + V(q) [\tilde{F}_+(q) + \tilde{F}_-(q)]},$$

where $\tilde{F}_{\pm}(q) = F_{\pm}(q) [1 - U(q)F_{\pm}(q)]^{-1}$. Within the same approximation the dynamical susceptibilities are obtained by replacing $F_{\sigma}(q)$ by $F_{\sigma}(q, \omega)$ $= -\sum_{k} [f(\epsilon_{k\sigma}) - f(\epsilon_{k+q,\sigma})](\epsilon_{k\sigma} - \epsilon_{k+q,\sigma} + \omega)^{-1}$ in Eqs. (3').⁷ The expression for $\chi_{mm}(q)$ is the same one that Schrieffer⁸ obtained. When $F_{+}(q) = F_{-}(q)$ = F(q), the susceptibilities reduce to the correct paramagnetic responses, i.e., $\chi_{mm}(q) = 2\mu_{\rm B}^2 F(q) [1 - U(q)F(q)]^{-1}$, $\chi_{ee}(q) = 2e^2 F(q) \{1 + [2V(q) - U(q)] \times F(q)\}^{-1}$ (Ref. 5), and $\chi_{em}(q) = \chi_{me}(q) = 0$.

In Figs. 1 and 2 we plot the responses as a function of the magnetization for parabolic bands with values for V(q), U(q), and $k_{\rm F}$ typical of Pd *d*-band electrons.⁹ For comparison, in Fig. 1(b),



FIG. 1. Comparison of (a) $\chi_{mm}(q)$ and (b) $\chi_{IKK}(q)$ as a function of the magnetization $M = [n_+(0) - n_-(0)][n_+(0) + n_-(0)]^{-1}$ for interacting *d*-band electrons typical of Pd. $N(0)U(q) = N(0)U(0)[1 + Bq^2/12k_F^2]^{-1} = 0.9[1 + 7q^2/12k_F^2]^{-1}$, $k_F = 0.5 \times 10^8$ cm⁻¹, and the effective mass of the electron is chosen to be eight times that of the free electron.

we have included the previously used $\chi_{IKK}(q)^6$ for the spin susceptibility. The difference between $\chi_{mm}(q)$ and $\chi_{IKK}(q)$ is practically negligible except for values of M close to 1. Thus the reanalysis of the neutron scattering in FePd alloys¹⁰ remains virtually unchanged except for a small increase from 3.0 to 3.3 $\mu_{\rm B}$ in the magnetic moment attributed to the Fe impurity.¹¹ $\chi_{mm}(q)$ of Eq. (3) correctly reduces to the Stoner susceptibility in the limit as $q \rightarrow 0$: $\chi_{mm}(0) = (4\mu_B^2)$ $\times [1/N_{+}(0) + 1/N_{-}(0) - 2U(0)]^{-1}$, where $N_{\pm}(0)$ are the densities of states of the spin-up and -down bands at the Fermi surface. The discontinuity of $\chi_{I \text{ KK}}(q)$ at q = 0 can be traced to the use of a δ -function interaction. In the spin split band the response to the wave-number-dependent magnetic field tends to accompany charge polarization, but the long-range part of the Coulomb interaction resists this electron concentration polarization. In the δ -function model this longrange part of the Coulomb interaction is neglect-



FIG. 2. The charge and spin response to a pointcharge potential as a function of the magnetization for the same band parameters used in Fig. 1. In part (b) for small M, the peak at $q/2k_{\rm F}=1$ can be traced to the infinite slope in the Lindhard functions F(q) at $q=2k_{\rm F}$.

ed in the original Hamiltonian.

For a point charge Ze the total screening charge is correctly obtained from the limit as $q \rightarrow 0$ of our susceptibility: $-4\pi Zeq^{-2}\chi_{ee}(q) = -Ze$, independent of the magnetization.

Although $\chi_{me} = \chi_{em} = 0$ for the paramagnetic state, in the ferromagnetic case these nondiagonal susceptibilities are as important as the usual diagonal susceptibilities. If we assume that the effect of adding an impurity can be represented typically by a charge Ze and spin S interacting with the conduction electrons via an exchange interaction J(s-d interaction), then the total spin polarization induced in the medium due to the charge and the spin are given, respectively, by $-Z \mu_{\rm B} [N_{+}(0) - N_{-}(0)] [N_{+}(0) + N_{-}(0) - 2U(0)N_{+}(0)]$ $\times N_{-}(0)^{-1}$ and $4(JS/N)\mu_{B}[1/N_{+}(0)+1/N_{-}(0)-2U(0)]^{-1}$, where for typical values of Z and S these two contributions are on the same order of magnitude. Recently neutron-diffraction experiments¹² and Mössbauer experiments¹³ revealed a rather complicated behavior for the spin and charge polarization around impurities in ferromagnetic metals and some attempts were made to analyze this behavior theoretically.² We believe our results offer a sound basis for this kind of analysis.

We would especially like to thank Professor J. R. Schrieffer for discussions related to χ_{nnn} (q). These discussions helped stimulate our present work.

†Supported by the U.S. Air Force Office of Scientific Research.

¹J. Friedel, Nuovo Cimento Suppl. <u>2</u>, 287 (1958). ²J. Kanamori, J. Appl. Phys. <u>36</u>, 929 (1968); I. A. Campbell and A. A. Gomes, Proc Phys. Soc. (London) <u>91</u>, 319 (1967); G. G. Low, Proc. Phys. Soc. (London) 92, 938 (1967).

³P. N. Zubarev, Usp. Fiz. Nauk <u>71</u>, 71 (1960) [translation: Soviet Phys.-Usp. 3, 320 (1960)].

⁴M. Watabe, Progr. Theoret. Phys. (Kyoto) <u>28</u>, 265 (1962).

⁵J. Hubbard, Proc. Roy. Soc. (London), Ser. A <u>243</u>, 336 (1957). Hubbard has discussed the effects of exchange processes on the dielectric constant $\epsilon(q, \omega)$ in the paramagnetic state in the same approximation as ours; in terms of our notation his result is $\epsilon(q, \omega) = 1$ + $[2V(q) - U(q)]F(q, \omega)$. In his paper Hubbard discusses why U(q) does not diverge as $q \rightarrow 0$, whereas V(q) is the unscreened Coulomb potential.

⁶T. Izuyama, D. J. Kim, and R. Kubo, J. Phys. Soc. Japan <u>18</u>, 1025 (1963).

⁷To calculate $\chi_{mm}(q, \omega)$ from the Kubo formula, for instance, Eq. (4.20) of Ref. 6 should be replaced by

$$[\theta_{k\sigma}(q), \mathcal{K}_{c}] = V(q) (n_{k+q,\sigma} - n_{k\sigma}) \sum_{l,\sigma'} \theta_{l\sigma'}(q)$$
$$-U(q) (n_{k+q,\sigma} - n_{k\sigma}) \sum_{l,\sigma'} \theta_{l\sigma}(q).$$

⁸J. R. Schrieffer, private communication. Expressions for the spin susceptibility taking into account the charge conservation have been discussed separately by J. R. Cullen, to be published; S. D. Silverstein, M. J. Rice, and L. M. Roth, to be published; and P. Martin, private communication.

⁹A. M. Clogston, Phys. Rev. Letters <u>19</u>, 583 (1967). For the actual model calculation we use a Lorentzian form for $U(q) = U(0) [1 + Bq^2/12k_F^2]^{-1}$. In Ref. 5, B was equal to 12.

¹⁰G. G. Low and T. M. Holden, Proc. Phys. Soc. (London) 89, 119 (1966).

¹¹D. J. Kim and B. B. Schwartz, Phys. Rev. Letters <u>21</u>, 1744 (1968).

¹²For a review see G. G. Low, J. Appl. Phys. <u>39</u>, 1174 (1968).

¹³G. K. Wertheim, V. Jaccarino, J. H. Wernick, and D. N. E. Buchanan, Phys. Rev. Letters 12, 243 (1964); M. B. Stearns, Phys. Rev. 147, 439 (1966).

CHEMICAL EFFECT ON THE HALF-LIFE OF U^{235m}

M. Nève de Mévergnies

Centre d'Etude de l'Energie Nucléaire-Studie Centrum voor Kernenergie, Mo-Donk, Belgium (Received 30 June 1969)

The half-life of the low-energy isomeric state of U^{235} produced by α decay of Pu^{239} was measured as a function of various metallic environments. The results show a correlation between the half-life and the average electronegativity of the host metal. In addition, effects due to impurity diffusion of U in metals at room temperature are observed.

The transition rate between two nuclear states is usually independent of the chemical or physical state of the sample. This behavior can eventually be violated,¹ e.g., when the transition energy between two nuclear levels of a given nucleus is exceptionally low: In this case, the transition proceeds by conversion in the outer electron shells of the atom, and a perturbation of these shells by chemical or physical means can alter the transition rate in a measurable way. This is