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HYPERFINE FIELD AT A POINT INTERSTITIAL IMPURITY IN FERROMAGNETIC NICKEL

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HYPERFINE FIELD AT A POINT INTERSTITIAL IMPURITY IN FERROMAGNETIC NICKEL*

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A simple model for the spin polarization around a charged interstitial impurity in ferromagnetic nickel is presented. It is based on screening of the charge by only s-electrons, while d-electrons, considered to be correlated and localized, are responsible, through an exchange mechanism, for the s-electron spin polarization. Agreement with recent experiments on μ^+ precession in Ni, as well as neutron diffraction data is satisfactory.

The hyperfine field experienced by an interstitial positive muon in ferromagnetic nickel has recently been measured[1,2] to be -0.66 kG at 77 K. In these measurements, a stopped muon is allowed to precess in the local field at the stopping site (presumed in this case to be the octahedral interstitial site), and this precession is monitored via positrons from the anisotropic decay of the μ^+. In what follows, an attempt is made to explain the sign and magnitude of the observed hyperfine field via a simple model.

The conduction electron contribution to the hyperfine field in a ferromagnet may be written[3]:

$$B_{hf} = \frac{-8\pi}{3} \mu_B \left[n^{\uparrow}(0) - n^{\downarrow}(0) \right] = \frac{-8\pi}{3} \mu_B \, n(0) \quad \zeta(0)$$

where $n^{\uparrow}(0)$ and $n^{\downarrow}(0)$ are conduction electron spin densities at the impurity,

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and $\zeta(0) = [n^{\uparrow}(0) - n^{\downarrow}(0)]/n(0)$. Using neutron diffraction, Mook and Shull[4] found that the moment density in pure ferromagnetic nickel has the form shown in Figure 1a. The unperturbed moment density at the octahedral interstitial site is:

$$-\mu_B\ [n_o^{\uparrow} - n_o^{\downarrow}] = -\mu_B\ \zeta_o\ n_o = -0.85 \times 10^{22}\ \mu_B/\mathrm{cm}^3 = -0.079\ \mathrm{kG}.$$

Consider first a "very naive" picture in which no screening of the impurity charge occurs. Then $n^{\uparrow\downarrow}(0) = n_o^{\uparrow\downarrow}$, giving $B_{hf} = \frac{8\pi}{3} \times (-0.079)$ kG $= -0.66$ kG, in perfect agreement with experiment. In order to explain the success of this very naive picture while taking account of screening, a way must be found to increase the charge density at the impurity without increasing the spin density.

It should be noted at this point that most band structure models[5-7] which consider both d- and s-electrons as itinerant cannot explain the existence of large regions in the crystal where the spin density is opposite to the majority spins. This is because the exchange splitting of all electrons tends to have the same sign, throughout the Brillouin zone. One notable exception is the work of Connolly[8], who through a self-consistent calculation obtains an s-electron exchange splitting opposite in sign to that of the d-states.

It is in any case probable that the negative moment density in the interstitial region of nickel (Fig. 1a) is due to the 4s-electrons and that the more localized, positive moment is due to the 3d-electrons. We carry this to the extreme in a simple model which treats the 3d-electrons as perfectly localized on the nickel cores and the 4s-electrons as forming a free electron gas. Each nickel core contributes 0.6 electrons to the 4s-band, giving a uniform unperturbed 4s-electron density $n_o = 4.9 \times 10^{22}\ \mathrm{cm}^{-3}$. A schematic diagram of the unperturbed spin densities according to our model is shown in Figure 1b.

The 4s-electrons move to screen the impurity, forming a screening cloud with a radius given roughly by the Fermi-Thomas screening length $r_s = 0.6$ Å, appropriate for this electron density. Since the impurity-nickel distance is 1.8 Å, all screening is by 4s-electrons in this model. Using the Lindhard expression for the free electron gas dielectric function[9], one finds the perturbed electron density at the impurity is $n(0) \sim 5\, n_o$.

A simple picture predicts that the screening cloud has the same proportion of spin up and spin down electrons as the unperturbed state. Since the very naive picture of no screening gives the correct hyperfine field, this direct proportionality hypothesis will give too large a field (by a factor 5).

Consider now the form of the total energy of the free 4s-electrons. This includes: kinetic energy, s-d exchange energy, s-s exchange energy, and correlation energy. Thus, the energy density in the unperturbed case is:

$$\varepsilon_o = \frac{1}{V} \, [E_{ke} + E_{sd} + E_{ss} + E_c],$$

where V is the volume of the sample. For the moment, we ignore the terms E_{ss} and E_c and break E_{ke} and E_{sd} into spin up and spin down components:

$$\varepsilon_o = A\, n_o^{5/3} \, [(1 + \zeta)^{5/3} + (1 - \zeta)^{5/3}] + \mu_B \, H_{sd} \, n_o \, \zeta.$$

Here, $A = 1.44 \frac{\hbar^2}{m}$, and $\zeta = \frac{n^{\uparrow} - n^{\downarrow}}{n_o}$. The first term is kinetic energy and comes from summing free electron states for the two spin orientations up to the Fermi level. In the second term, it is assumed that the effect of the s-d exchange interaction can be approximated by a Zeeman interaction with an effective, uniform, exchange field, H_{sd}.[8]

The equilibrium value of the spin density, found by miminizing ε_o with respect to ζ, is $\zeta_o \propto -H_{sd}\, n_o^{-2/3}$. For the unperturbed case, $\zeta_o = -0.85/4.9 = -0.17$; the value of H_{sd} is fixed to give this spin density. One finds that $H_{sd} \sim -10^8$ G, a field of the order of normal Weiss fields (but with the opposite sign).

The point impurity is now introduced, and we assume that its Coulomb field does not affect the magnetic terms in the hamiltonian. We also assume a "local" approximation, i.e., ε_o goes to $\varepsilon(x)$ as n_o goes to $n(x)$ and ζ_o to $\zeta(x)$; the kinetic and magnetic energies at the point x depend only on the charge and spin densities at point x. The charge density $n(x)$ is determined by Coulomb effects, with a negligible magnetic contribution.

It is only the region near the impurity that is of interest, so we minimize $\varepsilon(0)$ with respect to $\zeta(0)$ to find the equilibrium spin density at the impurity. We may then investigate how $\zeta(0)$ changes with $n(0)$ (i.e., what the relation is between the charge and spin densities at the impurity). It is convenient to define two quantities: $\rho_s = \frac{n(0)\,\zeta(0)}{n_o\,\zeta_o}$ and $\rho_q = \frac{n(0)}{n_o}$, the relative spin and charge densities, which compare the perturbed and unperturbed states. The success of the very naive picture implies that the true value of ρ_s must be close to 1, and the calculation of the Lindhard screening implies that ρ_q must be ~ 5. From a plot of the calculated relationship between ρ_s and ρ_q (Fig.2), it is clear that the model is a considerable improvement over the direct proportionality hypothesis ($\rho_q = \rho_s$). When the effects of s-s exchange and correlation[10] are taken into account according to generally accepted schemes $\{E_{ss} \propto n_o^{4/3} [(1 + \zeta)^{4/3} + (1 - \zeta)^{4/3}]$ and $E_c \propto n_o [(1 + \zeta) \log (1 + \zeta) + (1 - \zeta) \log (1 - \zeta)]\}$ the agreement improves slightly (see Fig. 2).

However, we have made a serious error in using the local approximation in the expression for the energy density in the perturbed state. The local approximation can only be valid when changes in charge density occur slowly over a typical electron wavelength $\lambda \sim 1/k_F \sim a$, where a is the lattice constant. But the charge density near the impurity changes drastically in a distance $r_s = 0.6$ Å $= a/6 < a$. Thus the local approximation is not valid, and one cannot have much confidence in the quantitative aspects of the calculation.

It is hoped, however, that the main theses of our argument will hold up to closer scrutiny. These are: 1) The neutron diffraction data correctly give the moment density, and the negative interstitial moment is from quasi-free 4s-electrons. This implies an antiferromagnetic s-d exchange interaction analagous to that found in rare earths[11]. We believe the μ^+ result and our simple theory support the neutron work. 2) The 3d-electrons participate only weakly in screening the impurity as if they were highly correlated on each Ni core, moving essentially as a localized unit. Such a high degree of correlation is evident[12] in NiO. 3) The kinetic energy increase accompanying a build-up of one spin orientation in the screening cloud keeps the spin density at the impurity low while the charge density increases.

An extension of the theory presented may be made to the case of Knight shifts seen by point impurities in normal metals simply by replacing H_{sd} by the known applied external field. Since ζ_o is proportional to this field, so is the hyperfine field, as in the case of Knight shifts. The results of a calculation of $\Delta B/B$ for various values of $n(0)$ is presented in Figure 3a. The rise at low densities found when s-s exchange and correlation are included is the well known exchange enhancement of the spin susceptibility.

Data of Knight shifts of μ^+ precession frequencies of Hutchinson <u>et al</u>[13] (Figure 3b) follows the general shape of the exchange enhanced theoretical curve.

Measurements of the local magnetic field at positive muons stopped in ferromagnetic iron[1,14] and cobalt[14] have recently been published. The case of iron is ambiguous due to non-zero dipolar fields and a complicated moment density[15] at the interstitial sites. The hyperfine field at the muon in ferromagnetic cobalt agrees qualitatively with a "no screening" picture[16].

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FIGURE CAPTIONS

FIG. 1. a) Magnetic moment density along the [100] direction in ferromagnetic Ni as measured[4] by neutron scattering. b) Schematic diagram of spin distribution according to a simple model.

FIG. 2. Calculated relationships between the relative charge and spin densities (ρ_q and ρ_s) at the impurity site. ρ_q and ρ_s are the charge and spin densities at the impurity normalized to the unperturbed, or impurity-absent, situation. The straight line $\rho_s = \rho_q$ represents the "direct proportionality" hypothesis, while the remaining curves are the result of model calculations involving the designated terms in the expression for the free electron energy E_{fe}.

FIG. 3. a) Calculated dependence of the impurity Knight shift ($\Delta B/B$) on the free electron charge density at the impurity n(0). The two curves represent calculations involving the designated terms in the expression for the free electron energy E_{fe}. E_z is the Zeeman energy of the free electrons in the externally applied field. b) Knight shift data[13] of μ^+ in various metals. Here, n(0) is the free electron density at the μ^+, calculated as described for Ni in the text.

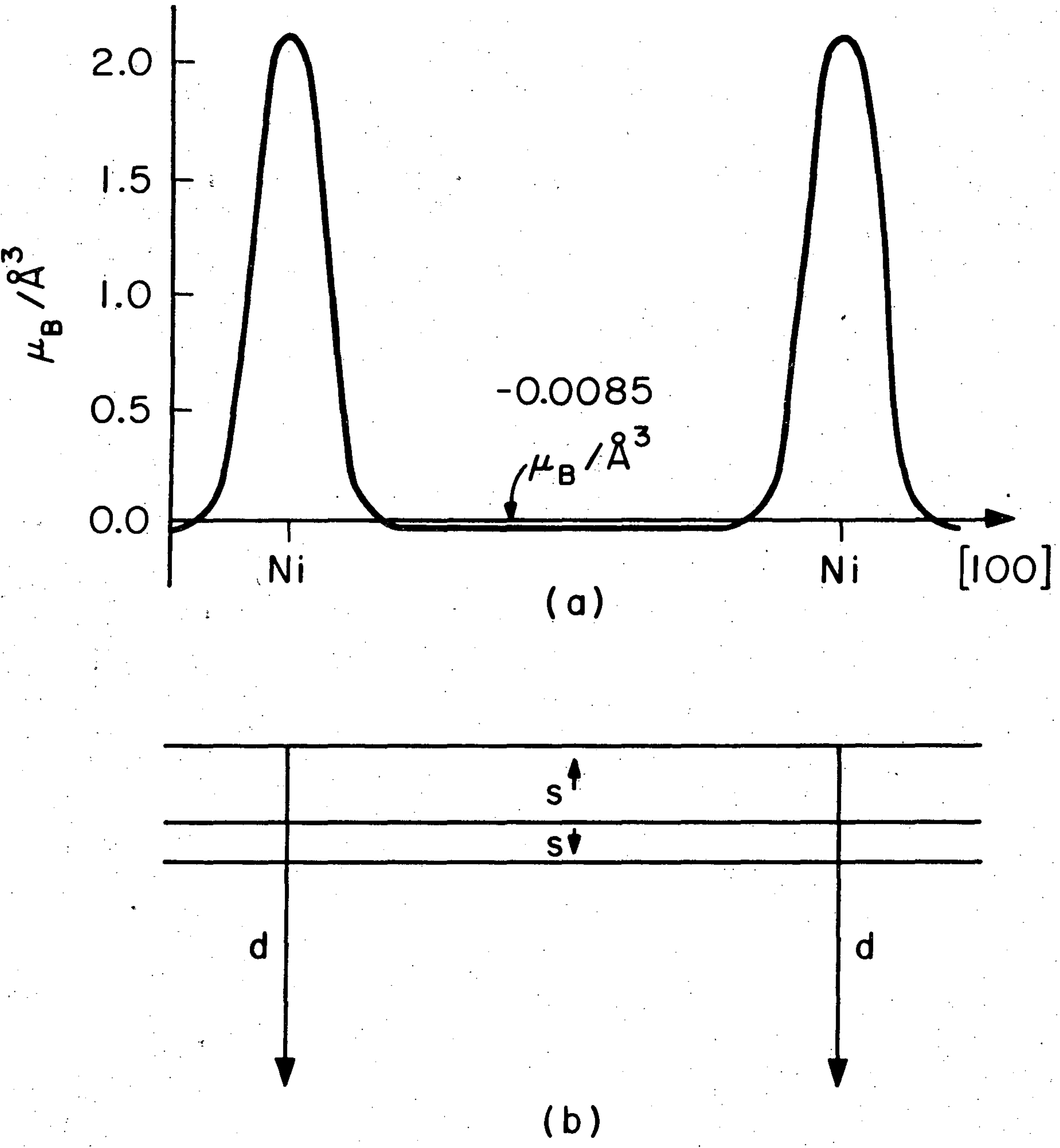

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Fig. 1

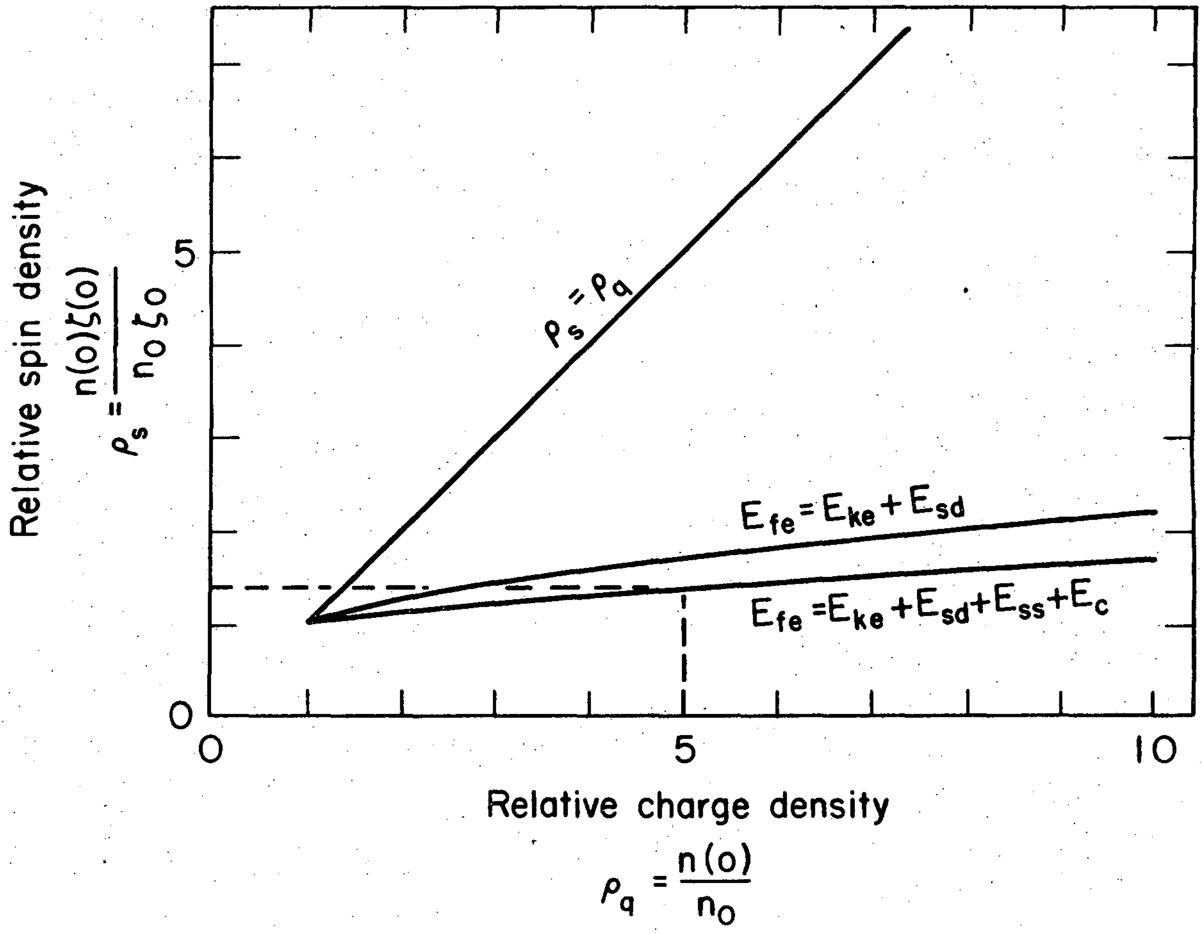
Relative spin density
$\rho_s = \frac{n(o)\zeta(o)}{n_o\zeta_o}$
0
5
0
5
10
Relative charge density
$\rho_q = \frac{n(o)}{n_o}$
$\rho_s = \rho_q$
$E_{fe} = E_{ke} + E_{sd}$
$E_{fe} = E_{ke} + E_{sd} + E_{ss} + E_c$
XBL 746-3443

Fig. 2

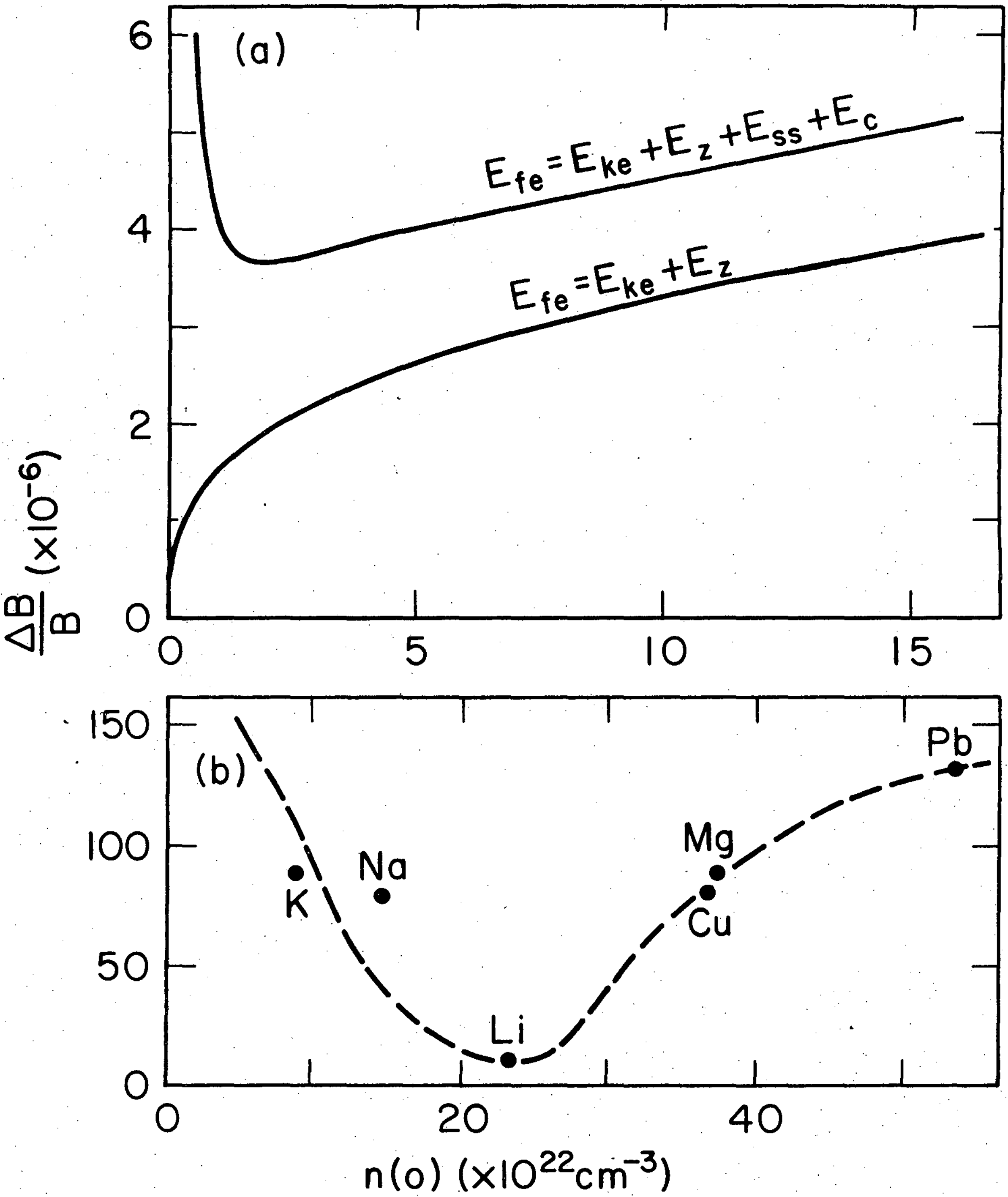

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Fig. 3

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