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We have found a smooth variation with atomic number of induced hyperfine magnetic fields at nuclei of nonmagnetic atoms dissolved in magnetic lattices. Rather than being generally negative (i.e., antiparallel to an external magnetizing field) as has been previously believed, the induced fields go systematically through zero and become positive in the 5p shell. We believe that this trend constitutes a connecting link between induced fields in metals (e.g., Cu in Fe) and in ionic ligands (e.g., F^- in MnF_2).

Samoilov et al[1] first showed that large hyperfine fields exist at diamagnetic impurities in ferromagnetic lattices. These fields have received considerable attention recently, although their origins are still not well understood. Theoretical treatments based on polarization of core and conduction electrons have successfully described hyperfine fields for magnetic atoms in ferromagnetic lattices[2], but no general extension to nonmagnetic atoms has been made. Two empirical rules[3] have emerged: (1) Induced fields at nonmagnetic atoms are negative, and (2) Induced fields are proportional in magnitude to the host's atomic moment. Two apparently isolated exceptions are now known. A positive, though very small, field was found for Sn in Ni[4], and recently Samoilov has given evidence for positive fields at Sb nuclei in iron and nickel[5].

In this letter we report more hyperfine fields and show that the positive fields mentioned above are not anomalous, but are part of a systematic trend.

We restrict our remarks to the two hosts iron and nickel. To investigate the systematic variation of hyperfine fields with atomic number we have studied Ru in nickel, and Cd and Te in both nickel and iron. We also use Kistner's results for Ru in Fe[6], the data of Samoilov et al[1] for In in Fe, and recent work from this Laboratory on Ag in Fe and in Ni[7].

Our Ru and Cd fields were measured by time-differential angular correlations in Ru^{99} and Cd^{111}.[8] Magnitudes only were obtained in the Cd experiments, because no polarizing field was used.

The most significant contribution to the systematization of fields was the determination of the hyperfine fields of Te in iron and nickel. Mössbauer spectra[9] of the 35.5-keV $3/2^+(M1)1/2^+$ transition in Te^{125}, using sources of Sb^{125} in Fe and Ni, showed a 3:2:1:1:2:3 pattern (Fig. 1a), established $\mu_{35.5}$ as positive (μ_0 = -0.88715 nm[10]) and gave the magnitudes of the hyperfine fields. Independent confirmation of the sign of $\mu_{35.5}$ was obtained by integral angular-correlation rotation measurements on the highly anisotropic 426-35.5 keV cascade, using a Ge(Li) detector for the 35.5-keV γ ray (Fig. 1b). The signs of the hyperfine fields were determined by angular-correlation rotation in polarized alloys.

These fields and others in this region of the periodic table are summarized in Table I. They show an interesting systematic trend, exhibited in Fig. 2. Hyperfine fields of the more metallic atoms Ru, Ag, Cd, and In are negative (where the sign is measured), as expected. They probably arise from contact hyperfine interactions through polarized conduction and core electrons. For Sn, Sb, and Te the 4d shell is full and the 5s and 5p shells are filling. We might expect the induced fields to become quite small after filling of the "magnetic" 4d shell and the 5s shell, but in fact the fields change sign and become quite large. We feel that there are a priori two rather distinct parameters with which this trend may be associated:

(1) filling of the 5p shell per se, and (2) the tendency in Sn-Sb-Te toward nonmetallic behavior. If p-shell filling is a crucial parameter we would expect the fields to follow the number of unpaired p electrons and probably to show a maximum near the middle of the 5p shell (Sb). As the p shell fills, the number of unpaired p spins must go nearly to zero. There might then be some semblance of a Slater-Pauling curve for induced fields in p shells. Already there is good evidence for such a curve in 3d series atoms dissolved in Fe[3], and some evidence for 4d series atoms. The mechanism for core polarization by p electrons would presumably be similar to that operative in nitrogen[11], although already for $Z \cong 50$ relativity effects may also be important. Koi and co-workers have indicated that hyperfine fields at nuclei of As ($4s^2 4p^3$) in MnAs and Sb in MnSb may be positive.[12]

The nonmetallic behavior of Sn, Sb, and Te may also be responsible for positive fields. Ionic configurations such as $Te^=$ ions are large (ionic radius = 2.2 A); Te 5s electrons should overlap with 3d electrons on neighboring Fe atoms. Ferromagnetic spin polarization of Te 5s electrons by Fe 3d electrons would produce a positive contact hyperfine field at the Te nucleus. Of course an ionic mechanism is not necessary; transferred hyperfine fields can arise by polarization through hybridized covalent bonds (still involving the 5s electrons) as well. We favor the 5s mechanisms because they can easily account for the magnitudes of the induced fields if the Te 5s electrons are polarized to the extent of a few percent[3]. Freeman and Watson have discussed similar mechanisms for F^- in transition-metal fluorides[13].

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Table I. Hyperfine Fields in KGauss for Atoms in Fe and Ni Hosts.

Atom Host	Ru	Ag	Cd	In	Sn	Sb	Te
Fe	500	-350(100)	348(10)	~250	-81(4)	+205	+620(20)
Ni	-178(7)	-108(30)	65.3(1.6)		+18.5(1.0)	~+70	+195(10)

FIGURE CAPTIONS

Fig. 1. a) Hyperfine Mössbauer absorption spectrum of 35.5 keV transition in Te^{125}.

b) Angular correlation of 426-35.5 keV cascade in Te^{125} normalized to the K_α x-ray intensity.

Fig. 2. Hyperfine fields for solute atoms in Fe (open circles) Ni (filled circles) hosts, vs. atomic number. Signs are known for all fields except Ru, Cd, In in Fe and Cd in Ni. Connecting lines are shown only to emphasize trends.

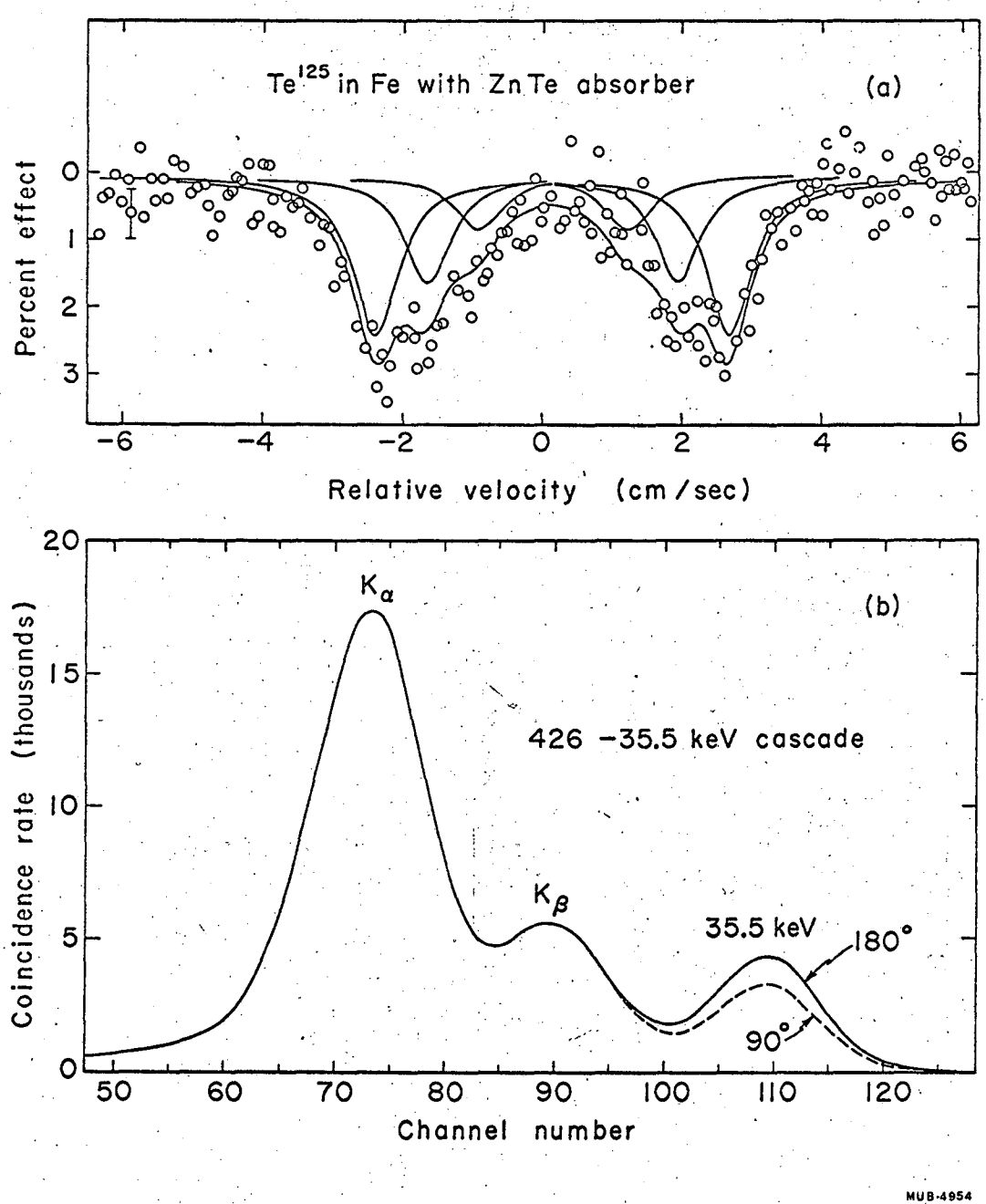
Te125 in Fe with ZnTe absorber
(a)
Percent effect
0
1
2
3
-6
-4
-2
0
2
4
6
Relative velocity (cm/sec)
(b)
426 -35.5 keV cascade
Kα
Kβ
35.5 keV
180°
90°
Coincidence rate (thousands)
0
5
10
15
20
50
60
70
80
90
100
110
120
Channel number
MUB-4954

Fig. 1.

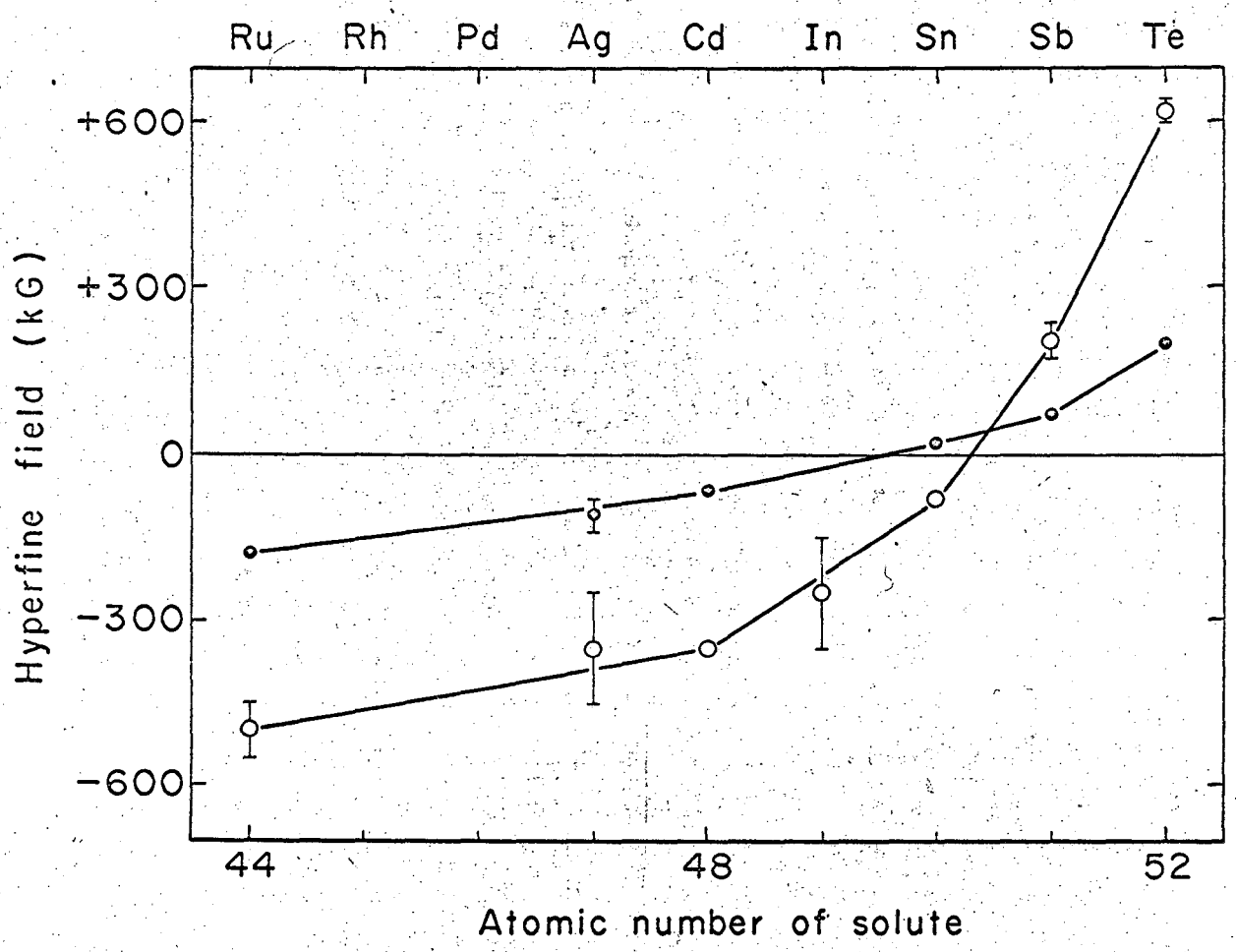
Ru
Rh
Pd
Ag
Cd
In
Sn
Sb
Te
+600
+300
0
-300
-600
Hyperfine field (kG)
44
48
52
Atomic number of solute
MUB-4953

Fig. 2.

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