

glass by the triad model with only unidirectional anisotropy provided that $T/T_g \ll 1$ and $\theta_r \approx 90^\circ$. At larger values of θ_r there is presently no description which encompasses all of the data though at exactly 180° the vector model with unidirectional and uniaxial anisotropy fits surprisingly well. Given the discrepancy between theory and experiment at large angles it may not be appropriate to infer any specific interpretations about the spin-glass state from measurements made under such conditions until further clarification.

We wish to thank Professor B. Halperin, Professor W. Saslow, and Professor C. Henley for helpful conversations. We thank Dr. A. Fert, Dr. H. Alloul, and Dr. F. Hippert for preprints of their work. This work was supported by the National Science Foundation under Grant No. DMR-80-07969.

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¹⁴At the lowest temperature the ESR data at $\theta_H = 180^\circ$ imply that $K^- \approx 0$. In this case the vector and triad models are indistinguishable. The data in Fig. 2 were taken at a temperature high enough to be able to distinguish between the two models yet low enough that the system still obeyed the triad model at small θ_H .

Enhanced Paramagnetism and Spin Fluctuations in Expanded Liquid Cesium

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(Received 8 November 1982)

¹³³Cs NMR has been used to probe the volume-dependent static and dynamic spin susceptibility of expanded liquid cesium. Data taken over the temperature range 55–1400°C cover the density range $1.03 < \rho < 1.92 \text{ g cm}^{-3}$. For $\rho \lesssim 1.5 \text{ g cm}^{-3}$, both the isobaric temperature dependence and the isothermal pressure dependence of the Knight shift change sign and the spin susceptibility becomes increasingly enhanced. Analysis of the nuclear relaxation rates at low densities indicates changes in the q -dependent susceptibility suggestive of a metallic antiferromagnet above its ordering temperature.

PACS numbers: 75.20.En, 71.45.Gm, 76.60.Cq, 76.60.Es

Liquid alkali metals, expanded by heating toward the liquid-gas critical point, exhibit interesting and unusual properties. Conduction-electron densities nearly five times lower than those of ordinary metals can be achieved before metallic properties give way to a metal-nonmetal transition close to the critical point.^{1,2} As elemental,

monovalent metals they closely resemble the hypothetical expanded lattice of hydrogen or alkali atoms considered by Mott³ in his original discussion of the metal-nonmetal transition.⁴ Although similar in some respects to dilute metals obtained by solution of donors in a nonmetallic host, e.g., heavily doped semiconductors, liquid-met-

al-ammonia solutions, alkali-metal-rare-gas mixtures, etc., expanded liquid alkali metals are free of theoretical complications due to electrons interacting with a host material.

The work of Brinkmann and Rice⁵ and recent theoretical studies of expanded hydrogen lattices⁶⁻⁸ predict unusual magnetic properties for low-density metals. The latter calculations, in particular, predict a metallic, antiferromagnetic ground state in a density range between the normal (paramagnetic) metal and the transition to an antiferromagnetic nonmetal. At high temperatures, enhanced paramagnetism might be expected in the low-density metallic range. This has been confirmed by Freyland's observation of increasing enhancements of the total molar and volume magnetic susceptibilities of cesium and rubidium as the density is reduced along the liquid-gas coexistence curve.⁹

In this Letter we describe the first application of nuclear magnetic resonance (NMR) to study an expanded monovalent metal. We have measured the ¹³³Cs Knight shift (K) and free-induction-decay lifetime (T_2^*) in liquid cesium up to 1400 °C at 120 bars and at pressures up to 900 bars at 300 °C and below. The minimum density achieved is about 56% that of liquid cesium at the melting point and corresponds to a value $r_s=7.0$. (The density parameter r_s is the radius, in Bohr radii, of a sphere equivalent to the volume per electron.) The data permit the following conclusions. First, enhancement of the total susceptibility at high temperatures is due predominantly to the effect of volume expansion on the conduction-electron-spin susceptibility. Second, fundamental changes in the magnetic properties begin to develop at remarkably high densities, i.e., at more than 90% of the density at the melting point. Finally, the q dependence of the generalized spin susceptibility $\chi(q, \omega)$ changes in a manner which is consistent with fluctuations toward an antiferromagnetic or spin-density-wave state at low density.

The high-temperature, high-pressure sample environments necessary for these measurements were achieved within an internally heated Be-Cu autoclave pressurized with argon gas. The high-temperature zone created by a three-element non-inductive heater was insulated from the autoclave by a layered structure formed from a roll of textured molybdenum foil. A cylindrical cell constructed from single-crystal sapphire contained the bulk liquid-cesium NMR sample. The pressure within the cell was automatically balanced

against the pressure of the surrounding argon by means of an external connecting capillary.

NMR measurements were made at a frequency of 9.7 MHz by using a coherent pulsed NMR spectrometer. Although the spin-lattice relaxation time (T_1) could be measured with a standard π - $\pi/2$ rf pulse sequence up to about 600 °C, it was necessary generally to measure the free-induction-decay lifetime (T_2^*) following a single pulse. After correction for the inhomogeneity of the magnetic field, we found T_1 equal to T_2 , the spin-spin relaxation time.¹⁰

The Knight shift is due to the local field associated with the static, uniform paramagnetic spin susceptibility

$$K \equiv \Delta H/H = \frac{8}{3} \pi \langle |\Psi(0)|^2 \rangle_F \Omega \chi'(0, 0), \quad (1)$$

where $\langle |\Psi(0)|^2 \rangle_F$ is the electron probability density at the nucleus averaged over all states at the Fermi energy, Ω is the atomic volume, and $\chi'(q, \omega)$ is the real part of the volume susceptibility. As the volume changes at constant temperature, simple normalization requirements imply $\langle |\Psi(0)|^2 \rangle_F \propto \Omega^{-1}$. However, studies of sodium under compression¹¹ indicate a weaker dependence, roughly $\Omega^{-0.6}$.

Our results for the ¹³³Cs Knight shift in expanded cesium are summarized in Fig. 1.¹² The most striking feature is the pronounced minimum near $\rho = 1.5 \text{ g cm}^{-3}$ in the variation of K with ρ at constant pressure. The sharp increase in K at low density corresponds to the rise in total suscepti-

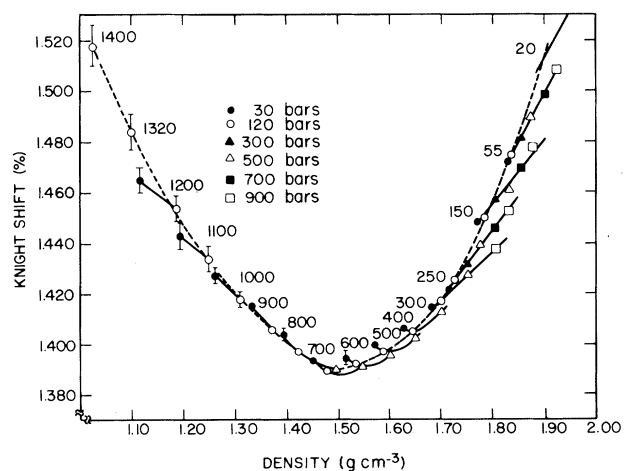


FIG. 1. Variation of the ¹³³Cs Knight shift with density at various temperatures (°C). The broken line is a guide to the eye connecting points along the 120-bar isobar. The shift variation in solid cesium at 20 °C represents data of Ref. 13.

bility observed by Freyland.⁹ At our lowest temperature, 55 °C, the isothermal variation of K with ρ , $(\partial \ln K / \partial \ln V)_{55} = -0.47 \pm 0.02$ agrees well with the value -0.46 measured by Benedek and Kushida¹³ at comparable densities for solid cesium at 20 °C. This demonstrates the relatively weak influence of long-range crystalline order on the volume-dependent magnetic properties. At 150 °C and 250 °C, $(\partial \ln K / \partial \ln V)_T$ agrees with the value $-\frac{1}{3}$ expected for a free-electron gas.

Beginning about 400 °C, the shift isotherms become nonmonotonic with pressure until, eventually, the pressure dependence becomes negative throughout our pressure range. The explicit (isochoric) temperature dependence $(\partial K / \partial T)_v$ is negative at high density, but tends to weaken at the lower densities. It is clear that the enhancement of the shift is mainly related to the reduced density and that temperature plays a secondary role. This is important for comparison with theory since calculations for expanded metals have so far been limited to consideration of volume expansion at absolute zero.

We consider now the nuclear relaxation rate and its relation to the Knight shift. The relaxation observed in liquid cesium is so strong that we need consider only relaxation due to the magnetic hyperfine interactions with conduction electrons and we neglect other mechanisms such as the nuclear dipole-dipole and electric quadrupole interactions. In this case the spin-lattice relaxation rate is governed by electron-spin fluctuations described by the low-frequency, transverse component of the imaginary part of the susceptibility $\chi''(q, \omega)$:

$$1/T_1 = \frac{64}{9} \langle |\Psi(0)|^2 \rangle_F^2 \gamma_n^2 k T \Omega^2 \omega_0^{-1} \times \int dq q^2 \chi_{+-}''(q, \omega_0), \quad (2)$$

where γ_n is the nuclear gyromagnetic ratio and ω_0 is the nuclear Larmor frequency. For noninteracting electrons, Eq. (2) yields the familiar Korringa rate¹⁴ which depends only on the temperature and the Knight shift,

$$(1/T_1)_{\text{Korringa}} = (4\pi k / \hbar) (\gamma_n / \gamma_e)^2 K^2 T. \quad (3)$$

More generally, electron-electron interactions¹⁵ or strong electron-ion scattering¹⁶ alter the q dependence of $\chi_{+-}''(q, \omega)$ with the result $\eta \equiv (1/T_1) / (1/T_1)_{\text{Korringa}} \neq 1$.

The effects of electron-electron interactions in simple metals such as the alkalis are usually discussed within the random-phase approximation

(RPA). The susceptibility takes the form

$$\chi(q, \omega) = \chi_0(q, \omega) / [1 - V(q)\chi_0(q, \omega)], \quad (4)$$

where $\chi_0(q, \omega)$ is the susceptibility in the absence of interactions and $V(q)$ is a q -dependent electron-electron interaction parameter. Defining a Stoner enhancement parameter $\alpha \equiv V(0)\chi_0'(0, 0)$ and $f(q) = V(q)\chi_0'(q, 0) / V(0)\chi_0'(0, 0)$ one has, at low frequency,

$$\chi'(0, 0) = \chi_0'(0, 0) / (1 - \alpha) \quad (5a)$$

$$\chi''(q, \omega) = \chi_0''(q, \omega) / [1 - \alpha f(q)]^2. \quad (5b)$$

Moriya¹⁵ has shown that enhanced susceptibilities of the form given by Eqs. (5a) and (5b) result in a deviation from the Korringa relation, Eq. (3), given by

$$\eta = K(\alpha) = (1 - \alpha)^2 / [1 - \alpha f(q)]^2. \quad (6)$$

The angular brackets denote an average over all q . Since $f(q)$ decreases at high q , $K(\alpha) < 1$ and $K(\alpha)$ tends to decrease as α increases [$K(1) = 0$].

The effects of electron-ion scattering on the Korringa ratio η have been observed for semiconducting liquid alloys.¹⁵ When the scattering becomes so strong that the conductivity σ falls below a value σ_0 corresponding to a mean free path λ equal to the interionic separation a , a diffusive model of electron transport predicts $\eta = \sigma_0 / \sigma > 1$. For cesium along the 120-bar isobar, the limit $\sigma \approx \sigma_0$ falls in the density range $0.6 \leq \rho \leq 0.8 \text{ g cm}^{-3/2}$.

Our experimental results for the ¹³³Cs relaxation rate at 120 bars are presented in Fig. 2 in the form η versus density. Near the melting point, $\eta = 0.61 \pm 0.02$ and, taking an explicit form for $K(\alpha)$ from the work of Shaw and Warren,¹⁷ we find an enhancement of the static, uniform susceptibility $(1 - \alpha)^{-1} = 1.82$. This is in reasonable agreement with the enhancements observed for the other alkali metals.¹⁷ However, it is immediately evident that the enhancement model characterized by Eq. (6) fails completely in the low-density region of enhanced $\chi'(0, 0)$ and K . In this range, we find that η increases as $\chi'(0, 0)$ and α increase whereas Eq. (6) predicts that η should decrease.

The failure of the conventional enhancement model indicates that fundamental changes in the q dependence of $\chi''(q, \omega_0)$ develop in the density range below about 1.4 g cm^{-3} . One possible explanation is that a gradual transition to diffuse transport begins at this density. However, at 1.4 g cm^{-3} $\lambda/a \sim 4$ and the density is roughly twice that expected for the onset of diffusive transport.

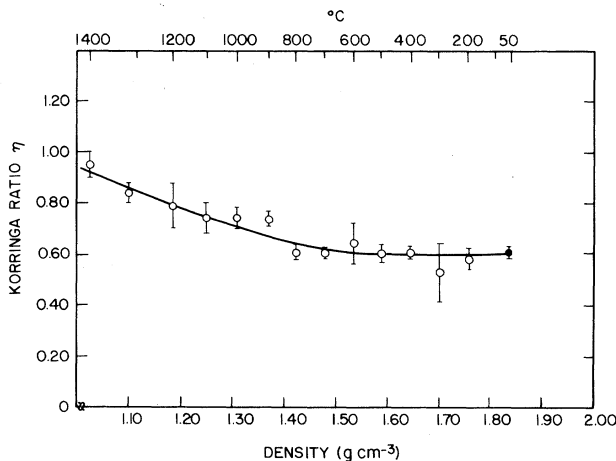


FIG. 2. Ratio (η) of the ^{133}Cs nuclear relaxation rate to the Korringa rate, Eq. (3), vs density under isobaric conditions at 120 bars. The upper scale gives the temperature scale in $^{\circ}\text{C}$. The solid point at maximum density was determined from direct T_1 measurements; open points were determined from free-induction-decay lifetime T_2^* , corrected for inhomogeneous broadening. The solid line is a guide to the eye.

Moreover, Hall-effect measurements by Even and Freyland¹⁸ indicate that nearly free electron (weak scattering) conditions hold throughout the range $1.1 \leq \rho \leq 1.9 \text{ g cm}^{-3}$. Finally, we point out that diffusive transport would not lead to the enhanced static susceptibility which is a pronounced characteristic of cesium in the low-density range.

An alternative and more likely explanation is the development of increased enhancement of $\chi''(q, \omega_0)$ around some value $q = Q_0 \neq 0$. A peak in $\chi''(q, \omega_0)$, and equivalently $\chi'(q, \omega_0)$, corresponds to fluctuations toward a spin-density-wave or antiferromagnetic state of wave vector Q_0 . The form of $\chi''(q, \omega_0)$ that we infer is essentially that considered by Hasegawa and Moriya¹⁹ who extended the RPA self-consistently to include enhancement by spin fluctuations, and by Usami and Moriya²⁰ who calculated the nuclear relaxation properties of an itinerant antiferromagnet above its ordering temperature. The complementary effect, reduction of η for a nearly ferromagnetic metal (TiBe), was recently reported by Alloul and Mihaly.²¹ At sufficiently low temperatures, expanded cesium would be a metallic antiferromagnet, but this is precluded by intervention of the liquid-gas phase separation.

In summary, we find that the magnetic properties of liquid cesium near the melting point can be closely approximated by those of an interacting electron gas with the usual Stoner enhance-

ment of the susceptibility, but a slight reduction in density is sufficient to introduce new effects. Presumably precursors of the eventual metal-nonmetal transition, these effects are an increased enhancement of the Knight shift and the static, uniform spin susceptibility, and a fundamental change in the q -dependent dynamic susceptibility. Although effects due to diffusive electron transport cannot be completely excluded, the overall similarity to a metallic antiferromagnet above its ordering temperature supports predictions of an antiferromagnetic ground state in the experimentally inaccessible range of low densities at low temperature.

The authors are pleased to acknowledge the contributions of J. Franz, W. Freyland, F. Hensel, and T. M. Rice with whom we discussed various aspects of this work.

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Decay and Regeneration of the Galactic Magnetic Field in the Presence of Magnetic Monopoles

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(Received 12 November 1982)

The resonant character of magnetic field damping by moving magnetic monopoles allows the field to survive indefinitely when the monopole plasma frequency is sufficiently large, even if the field is immersed in a conducting plasma. Dynamo growth of galactic field can still occur on time scales $\sim 10^7$ – 10^8 yr in the presence of a halo composed of monopoles. A linear stability analysis suggests that a monopole's mass is $m \sim 10^{18}$ GeV/ c^2 and that the monopoles should have an anisotropic flux $\sim 1 \text{ m}^{-2} \text{ yr}^{-1} \text{ sr}^{-1}$.

PACS numbers: 98.40.Bs, 14.80.Hv, 52.35.Hr, 98.50.Lh

Cabrera¹ has reported a possible candidate for a free magnetic monopole consistent with a Dirac charge $|g| = e/2\alpha_F \cong 3 \times 10^{-8}$ esu. Certain grand unification schemes² suggest that such monopoles should have a mass $m \sim 10^{16}$ GeV/ c^2 . (Hereafter mass in units of 10^{16} GeV/ c^2 will be denoted by m_{16} .) When applied to models of the very early universe,³ these theories indicate the possible existence of a large space density of free monopoles. One interesting possibility is that these monopoles could be the dominant mass contribution to the dark halo of our galaxy and of other spiral galaxies, which may be the origin of the observed stability of spiral galaxies' disks.⁴ Such stabilizing influence requires $M_{\text{halo}}/M_{\text{galaxy}} \gtrsim 1$, and demands the existence of some nonluminous form of matter in the halo of our own galaxy with mass density⁵ $\rho_h \gtrsim 10^{-24}$ g cm⁻³ and velocity dispersion $c_m \sim 200$ km/s. (Velocity in units 200 km/s are denoted by the subscript 200.) If the halo is composed of grand unified theory (GUT) monopoles, the expected flux at the earth would then be $\gtrsim 3[\rho_h/(10^{-24} \text{ g cm}^{-3})]m_{16}^{-1} \text{ m}^{-2} \text{ sr}^{-1} \text{ yr}^{-1}$, which is not inconsistent with Cabrera's event being a monopole of galactic origin.

However, several authors^{6,7} have argued that such high fluxes are inconsistent with the persistence of the galactic magnetic field, with estimates for upper bounds on the monopole flux $\sim 10^{-5} \text{ m}^{-2} \text{ yr}^{-1} \text{ sr}^{-1}$. We show that persistence of the galactic magnetic field is consistent with higher fluxes of monopoles, provided that the monopole plasma frequency exceeds a certain lower bound. We also show that dynamo activity⁸ in the interstellar medium can regenerate the magnetic field in the presence of a monopole-dominated halo, at a rate comparable to the orbital angular velocities of monopoles and stars in the halo and of stars and gas in the disk, $\sim (3 \times 10^7 \text{ yr})^{-1}$.

We expect a monopole halo to form a nonrotating,⁹ quasineutral, collisionless system with an approximately Maxwellian velocity distribution of dispersion speed c_m because of the effects of violent relaxation during galaxy formation.¹⁰ The monopole plasma frequency $\omega_{pm} = (4\pi g^2 N_m/m)^{1/2}$ is related to the angular frequency of the galaxy by

$$\left(\frac{\omega_{pm}}{\Omega_k}\right)^2 = \frac{g^2}{m^2 G} \left(\frac{M_m}{M_g}\right) = b \left(\frac{M_m}{M_g}\right), \quad (1)$$